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Copy of SSA Postcard submitted June 10, 2004. (1 pg.)
Excerpt from "Handbook of Powder Science and Technology" (67 pgs.)

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Firm or Individual	Peter Giancana, 44,706
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Serial Number: 09/932,060

Group Art Unit: 3732

Examiner: DOAN, R.

Filed: August 17, 2001

For: Case For Presenting And Using Cosmetic Powders

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

A Final Office Action was issued on October 5, 2004. On page six of the Office Action the Examiner remarked that in the previous response the applicant failed to provide copies of a reference that the applicant cited to support the his position.

As can be seen on the enclosed copy of the Return Postcard, item 5 reads, " Excerpt "Handbook Of Powder Science and Technology" (67 pgs.)". This postcard was stamped by the OIPE on June 14, 2004, indicating that the reference material was received at the Patent Office.

On December 20, 2004, the applicant contacted the Examiner to discuss. The Examiner asked for this submission, containing evidence of submission (i.e. a copy of the return postcard) and a re-submission of the 67 page reference. The Examiner indicated that the applicant's previous reply to non-final office action would be reconsidered and the present Final Office Action withdrawn for now.

Respectfully submitted,

Dated: DEC 21, 2004

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01.40 Gubernick 09/932060

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HANDBOOK OF POWDER SCIENCE & TECHNOLOGY

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edited by
Muhammad E. Fayed
Lambert Otten

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TO

My Late Parents,
Fat-Hia Hitata
Al-Sawi Fayed

My Wife Carolyn
and my children
Mark and Susan Otten

All of whom have given us far too much without reservation

CONTENTS

PREFACE TO THE SECOND EDITION

Since the publication of the first edition of *Handbook of Powder Science and Technology*, the field of powder science and technology has gained broader recognition and its various areas of interest have become more defined and focused. Research and application activities related to particle technology have increased globally in academia, industry, and research institutions. During the last decade, many groups, with various scientific, technical, and engineering backgrounds have been founded to study, apply, and promote interest in areas of powder science and technology. Many professional societies and associations have devoted sessions and chapters on areas of particle science and technology that are relevant to their members in their conferences and career development programs. Two of many references may be given in this regard; one is the recent formation of the Particle Technology Forum by the American Institute of Chemical Engineers. The second reference is the intensified effort given by the American Filtration and Separation Society to define the areas of particle and particle fluid science and technology with the objective to promote the inclusion of courses on these topics at American universities, for undergraduate and graduate ... Canada, and Australia have increased teaching, research, and training activities in areas related to particle science and technology.

In addition, it is worth mentioning the many books and monographs that have been published on specific areas of particle, powder, and particle fluid by professional publishers, technical societies and university presses. Also, to date, there are many career development courses given by specialists and universities on various facets of powder science and technology.

Taking note of all these developments, the editors of this second edition faced the need for evaluating and reorganizing, as well as updating and adding to the content of the first edition. In this edition, topics are organized in a logical manner starting from particle characterization and fundamentals to the many areas of particle/powder applications. Comprehensive upgrade of many of the first edition chapters were made and three more chapters were added: namely pneumatic conveying, dust explosion, and fire hazard and health hazard of dust.

The extent to which we have succeeded may be judged from the authors contributions and

6 Size Enlargement by Agglomeration*

Wolfgang Pietsch

CONTENTS

6.1 INTRODUCTION

6.1.1 Definition of Size Enlargement by Agglomeration

Size enlargement by agglomeration is a unit operation of mechanical process technology[1] (Fig. 6.1). This field deals with the transport phenomena and changes of state of particulate matter which in most cases is solid but can also be liquid (droplets) and, in a few special cases, gaseous (microencapsulated). The unit operations of mechanical process technology can be differentiated by the processes of separation and combination with and without change of particle size.

Size enlargement by agglomeration as a unit operation of mechanical process technology is characterized by the structure of the enlarged particles in which, contrary to, for example, crystals or particles obtained by solidification of melt droplets, the shape and size of the original particles are still distinguishable. This offers both advantages and disadvantages [redacted]

*References are listed at the end of sections 6.1 through 6.6.

Without change of Particle Size	Mechanical Separation (Filters, Classifiers, Screens, Sifters)	Powder Mixing and Blending	Partical and Bulk Material Characterization (Size, Distribution, Shape, Volume, Surface Density, Mass, Porosity, Moisture Content, etc.)
With change of Particle Size	Size Reduction (Crushing and Grinding)	Size Enlargement by Agglomeration	
	Transport and Storage of Bulk Materials		

Figure 6.1. The unit operations of mechanical process technology and associated techniques.

... solids or, respectively, by the negative capillary pressure of a liquid filling the ... (Fig. 6.2).

6.1.2 Properties of Fine Particles

Table 6.1 shows some important characteristics of materials and disperse systems that depend on particle size.[2]

For single particles the characteristics describing quality usually improve as particle size decreases. In particular, the chemical, physical, and mineralogical homogeneity increases. Those characteristics that critically depend on uniformity of structure improve also. For example, all real solids have an imperfect structure; during loading stress concentrations occur at the structural defects that may cause

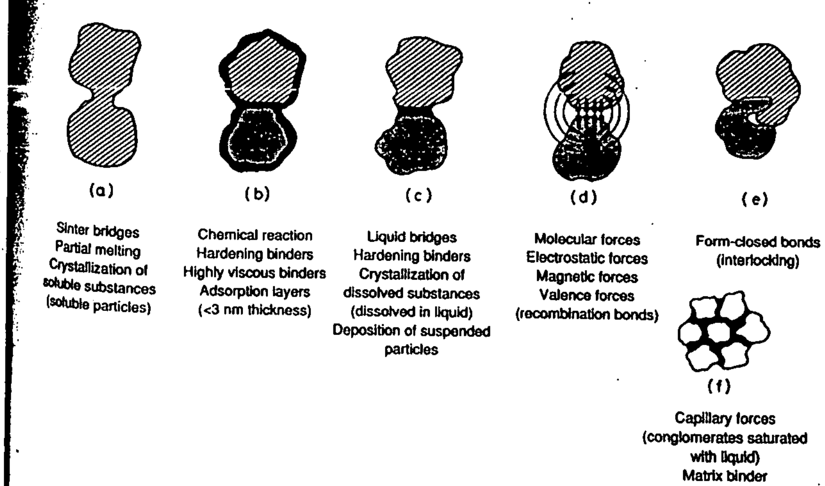

Figure 6.2. The binding mechanisms of agglomeration.

Table 6.1. Influence of Particle Size on some Important Characteristics of Materials.[2]

A.	Characteristics of Single Particle	...with decreasing particle size
A.1.	Homogeneity	Increasing...
A.2.	Elastic–plastic behavior	Increased ductility...
A.3.	a. Probability of breakage	Decreasing...
	b. Strength	Increasing...
A.4.	a. Wear	Decreasing...
	b. Resistance to mechanical surface treatment	Increasing...
A.5.	Characteristics resulting from the competition between volume and surface-related forces	Increasing...
A.6.	Vapor pressure, solubility, reactivity	Increasing...
A.7.	Optical characteristics	Increasing...
B.	Characteristics of Particle Collectives	
B.1	Bulk density (space-filling behavior)	First increasing then decreasing...
B.2.	Rheological behavior	Increasing...
B.3.	Flow characteristics, flowability (of particles)	Decreasing...
B.4.	Mixing characteristics	First increasing then decreasing...
B.5.	Separability	Decreasing...
B.6.	Wettability	Decreasing...
B.7.	Capillary pressure (system: solid/liquid)	Increasing...
B.8.	Agglomerate strength	Increasing...
B.9.	Fluid flow characteristics	
	a. Flow through pores (in particle collectives)	Decreasing...
	b. Resistance to fluid flow	Increasing...
	c. Ease of fluidization	First increasing then decreasing...
B.10.	Thermal characteristics	Increasing...
B.11.	Ignition behavior and explosiveness	Increasing...
B.12.	Taste standards	Increasing...
B.13.	Optical characteristics	Extinction, diffuse reflection

breakage. With decreasing particle size the probability of imperfections diminishes, resulting in a reduced risk of breakage and therefore higher strength. At the same time, the possibility for irreversible deformation increases with decreasing particle size. For example, limestone or quartz, with particle size of less than 10 μm and 3 μm, respectively, deforms plastically before breakage begins.

On the other hand, problems associated with mechanical processing and handling of particle systems increase with decreasing particle size mostly due to natural, undesired agglomeration including such phenomena as caking, bridging, build-up, etc.

Controlled or desired agglomeration may improve the characteristics of fine particle systems.

6.1.3 Desired and Undesired Agglomeration[3,4]

During production and processing of solid matter in disperse systems, adhesion phenomena become more and more important with decreasing particle size, causing aggregation, agglomeration, coating, caking, and build-up. The critical particle size is approx. 100 μm, but it is also possible that much coarser particulate matter may be affected if a sufficiently

large fraction of finer particles is present or if specific binding mechanisms become effective.

Adhesion of finely divided material takes place during all operations of mechanical process engineering and can be either desired or undesired. Table 6.2 provides a compendium.

Adhesion during *grinding* is always undesirable because it diminishes the grinding effect, lengthens the grinding time, and increases the energy requirement. In some mills an equilibrium between size reduction and size enlargement sets in at a certain fineness and can be avoided only by the addition of dispersion agents or the application of another comminution method.

During *mechanical separation* agglomeration is undesirable if products must be classified according to particle size or composition. Only in *flotation cells* or *wet classifiers* a "selective flocculation" may be advantageous. Particle aggregation is always desirable during precipitation, thickening, filtration, and clarification, because the increased mass of agglomerates improves separation efficiency.

During analytical separation (*particle size analysis*) any agglomeration is prohibitive and must be avoided at any cost.

The quality of *mixing of solids* can be considerably impaired by undesired agglomeration. Existing or newly formed aggregates are

Table 6.2. Review of the Occurrence of Desired and Undesired Agglomeration Phenomena in Mechanical Process Engineering.

UNIT OPERATION	PROCESS	AGGLOMERATION UNDESIRABLE	AGGLOMERATION DESIRABLE
Comminution	Dry grinding	+	–
	Wet grinding	+	–
Separation	Screening, sieving Classifying	+	–
	Sorting	+	(+)
	Flotation	+	(+)
	Dust precipitation	(–)	+
	Clarification, thickening	(–)	+
	Particle size analysis	+ +	–
Mixing	Dry mixing	+	–
	Wet mixing	+	+
	Stirring	+	(+)
	Suspending Dispersing	+	(+)
	Fluidized bed	+	+
Particle size enlargement	Agglomerating Briquetting Tabletting Granulating Pelletizing Pelleting Sintering	(+)	+ +
Conveying	Vibratory conveying	+	–
	Pneumatic conveying	+	–
Storage	Silos, hoppers	+	–
	Stockpile	+	–
Batching, Metering		+	–
Drying		+	+

...ormally destroyed by suitable mixing tools or by vigorous movements in the mixer. On the other hand, powder mixtures often tend to segregate during handling and storage; then, a controlled agglomeration of the final mix may be desirable prior to further processing.

Because fine powders possess a large bulk volume, generate dust, and exhibit unfavorable transport, storage, and feeding characteristics, their particle size is sometimes enlarged by *agglomeration*. In this case adhesion is desired and is systematically promoted. In some cases it is necessary to further treat the agglomerate with "anticaking" compounds to avoid clustering during storage.

Agglomeration and adhesion of fine particles are particularly annoying during *Transport, Storage*, and *feeding*. Conglomerates can result in clogging or feeders, prevent discharge from silos, and cause incorrect metering. The prevention or destruction of such conglomerates often requires considerable technical efforts.

Agglomeration can also play an important role in thermal unit operations. For example, if a liquid in the pores of a bulk mass contains dissolved substances that crystallize during *drying*, solid bridges may build up between the particles. Such bonding is often undesirable and must be destroyed by "deagglomeration." In other instances this method is used for "curing" a wet agglomerate, producing a stable granular material that is better suited as an intermediate product.

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1. W. Pietsch, *Size Enlargement by Agglomeration*. John Wiley & Sons/Salle + Sauerländer, Chichester, UK/Aarau, Switzerland (1991).
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6.2 AGGLOMERATE BONDING AND STRENGTH

6.2.1 Binding Mechanisms

To obtain agglomerates from [redacted]

6.2.1.1 Solid Bridges

Fig 6.2 a

At elevated temperatures, solid bridges may develop by diffusion of molecules from one particle to another at the points of contact ("sintering"). Heat can be introduced from an external, secondary source or created during agglomeration by friction and/or energy conversion. Solid bridges can also be build up by chemical reaction, crystalization of dissolved binder substances, hardening binders, and solidification of melted components.

6.2.1.2. Interfacial Forces and Capillary Pressure at Freely Movable Surfaces

Figs. 6.2 f

Capillary pressure and interfacial forces in liquid bridges can create strong bonds that disappear if the liquid evaporates and no other binding mechanism take over.

Table 6.3. Binding Mechanisms of Agglomeration.[1]

1. Solid bridges
2. Interfacial forces and capillary pressure at freely movable liquid surfaces
3. Adhesion and cohesion forces at not freely movable binder bridges
4. Attraction forces between solid particles
5. Form-closed bonds (interlocking)

...1.3 Adhesion and Cohesion Forces in Not ...ly Movable Binders

Figs. 6.2 b,c

...hly viscous bonding media such as tar and ...er highly molecular organic liquids can form ...ds very similar to those of solid bridges. ...n adsorption layers are immobile and can ...tribute to the bonding of fine particles ...der certain circumstances.

...1.4. Attraction Forces Between Solid ...ticles

Fig. 6.2 d

...e typical short-range forces of the van der ...als, electrostatic, or magnetic type can ...se solid particles to stick together if they ...roach each other closely enough. Decreas... particle size clearly favors this mechanism. ...freshly created surfaces after breakage, free ...lence forces are momentarily present which, at certain conditions, may recombine, forming strong bonds.

...2.1.5. Form-Closed Bonds

Fig 6.2 e

Fibers, little platelets, or bulky particles can interlock or fold about each other, resulting in "form-closed" bonds.

Another classification into only two groups[2] distinguishes between the presence of material bridges between the primary particles in the agglomerate and attraction forces (Fig. 6.3).

6.2.2. Theory of Agglomerate Bonding and Strength

[redacted] For the determination of agglomerate strength, real stresses are often simulated experimentally. In addition to the usually applied crushing, drop, and abrasion tests, methods for the determination of impact, bending, cutting, or shear strength are employed. All values obtained by these methods are strictly empirical and cannot be predicted by theory, since it is not known which stress component causes the agglomerate to fail. For the same reason, the experimental results from different methods cannot be compared with each other.

Therefore, Rumpf[1] proposed to determine the tensile strength of agglomerates. It is defined as the tensile force at failure divided by the cross-section of the agglomerate. Because with high probability failure occurs as the result of the highest tensile stress in all stressing situations, this proposal is justified. Moreover, the tensile strength can be approximated by theoretical calculations.

[redacted]

1. [redacted] Fig. 6.2 b,c
2. [redacted] Fig 6.2 f
3. [redacted] Figs. 6.2a,d,e

6.2.2.1 [redacted]

If the pore volume of the agglomerate is completely filled with a stress-transmitting substance, for example, a hardening binder, three strength components must be distinguished:

1. σ_{tc} (pore volume strength) = strength of binder substance
2. σ_{ta} (grain boundary strength) = strength caused by adhesion between binder and solids
3. $\sigma_{t(1-\epsilon)}$ = strength of the solids forming the agglomerate.

The relatively lowest strength component determines the agglomerate strength. If the pore volume strength or, respectively, the strength of the solids forming the agglomerate are the determining factors and if they are everywhere the same, then the cross-section of the respective material defines the agglomerate strength. A theoretical approximation is possible using the same assumptions as described below for solid bridges between parti...

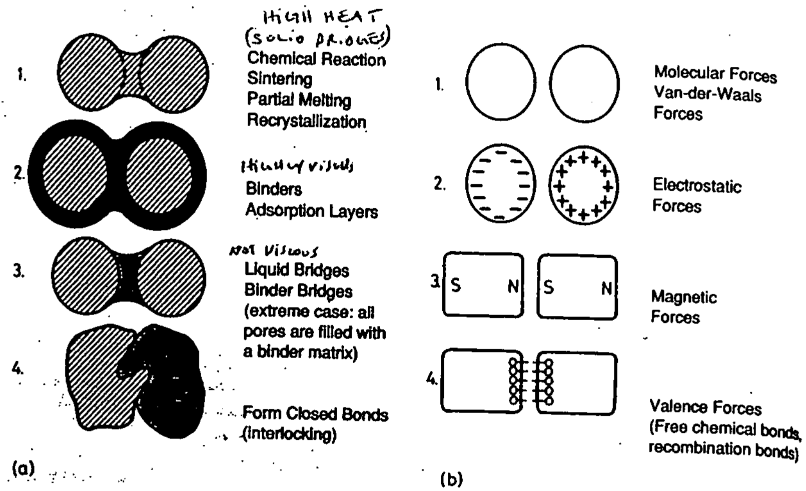


Figure 6.3. Alternative classification of the binding mechanisms.[2]

cles. If the agglomerate strength is caused by the grain boundary strength, it can be approximated by calculating the adhesion forces (see Section 6.2.2.4).

6.2.2.2 *Maximal Tensile Strength If the Pore Volume of the Agglomerate Is Filled with a Liquid*

If the entire pore volume of the agglomerate is filled with a liquid such that concave minisci are formed at the agglomerate surface a negative capillary pressure p_c develops in the interior. Because the membrane forces at the surface are negligibly small in relation to the capillary pressure, the tensile strength σ_{tc} of the agglomerates filled with a liquid can be approximated by the capillary pressure:

$$\sigma_{tc} \approx p_c \tag{6.1}$$

Assuming that the pore diameter is characterized by the mean half-hydraulic radius of the pore system, further assuming complete wetting and spherical monosized particles, the following equation is obtained:

$$\sigma_{tc} \approx p_c = a' \cdot \frac{1-\epsilon}{\epsilon} \cdot \alpha \cdot \frac{1}{x} \tag{6.2}$$

Therefore, the maximal tensile strength of agglomerates filled with a liquid is proportional to a porosity function $(1-\epsilon)/\epsilon$ and the surface tension of the liquid α; it is inversely proportional to the grain size x of the particles forming the agglomerate. The factor a' has a value between 6 and 8.

To correctly describe the capillary pressure, and thereby the tensile strength, a function of the wetting angle $f(\delta)$ would have to be included in the above formula. This function equals 1 if the liquid completely wets the solid.

Normally the particles forming agglomerates are not monosized and are irregularly shaped. Comparisons between experimental results and the theory showed that a mean grain size x_0, the surface equivalent diameter calculated from the specific surface area of the actual particle, describes the relations well.[3]

6.2.2.3 *Maximal Tensile Strength If Binding Forces Are Transmitted at the Contact and Coordination Points*

The model used for agglomerates, the strength of which is caused by solid bridges, assumes that the entire solid binder material is uniformly distributed at all contact and coordination points and, there forms bridges with constant strength σ_B. Then, the relative cross-sectional area of that material defines the agglomerate strength. In a random packing the cross-sectional area of one component (area porosity ϵ_a) is approximately equal to the relative volume of that same component (i.e., volume porosity $\epsilon_v = \epsilon_a = \epsilon$). Thus, the tensile strength σ_{tB} of agglomerates with solid bridges can be approximated by:[4]

$$\sigma_{tB} = \frac{M_B}{M_p} \cdot \frac{\rho_p}{\rho_B} \cdot (1-\epsilon) \cdot \sigma_B = \psi_B \cdot \epsilon \cdot \sigma_B \tag{6.3}$$

where M_B is the mass of the bridge building solid, M_p the mass of the particles forming the agglomerate, ρ_B and ρ_p the density of the respective solids, $(1-\epsilon)$ the relative unit volume of the solid, σ_B the tensile strength of the bridge building solid, ϵ the specific void volume porosity of the agglomerate, and ψ_B the saturation, that is, the fraction of the void volume filled with the bridge building material.

Equation (6.3) is valid only if, in addition to the restrictive assumptions mentioned above, failure occurs only through solid bridges. In reality all these conditions are never fulfilled. Particularly the uniform distribution and the constant strength of the binder material are seldom realized.

Often, the strength is caused by adhesion forces A acting at the coordination points of the particles forming the agglomerate. Based on statistical considerations Rumpf[5] developed a general formula for the tensile strength of such agglomerates. Assuming that the particles forming the agglomerate are monosized and spherical the tensile strength σ_t can be approximated by:

$$\sigma_t = \frac{1-\epsilon}{\pi} \cdot k \cdot \frac{A}{x^2} \tag{6.4}$$

where ϵ is the specific void volume (porosity) of the agglomerate, $\pi = 3.14$, k the average coordination number and x the size of the particles forming the agglomerate.

After a small correction[5] Eq. (6.4) can also be applied for nonspherical particles. Then the estimated elementary tensile strength σ_{to} becomes:

$$\sigma_{to} = (1-\epsilon) \cdot k \cdot A / O_p \tag{6.5}$$

with O_p the particle size. Equation (6.5) is valid for agglomerates formed by approximately isodisperse, convex, and monosized particles. With the third moment M_{30} of the number density distribution $n(x)$ and a shape factor f_0, a formula can be derived that is valid for distributions of similar, approximately isometric, and convex particles:

$$\sigma_{te} = \frac{1-\epsilon}{f_0 M_{30}} \int_0^\infty k(x) \cdot A[x, n(x)] \cdot x \times n(x)\, dx \tag{6.6}$$

This equation can be integrated only if the relationships are known between coordination number and particle size $k(x)$ as well as between adhesion force and particle size and distribution $A[x, n(x)]$. In most instances this is not the case. To measure σ_{te}, an agglomerate free of cracks must be uniformly stressed by tensile forces. This requires very sophisticated methods and experimental care (see Section 6.2.3).

6.2.2.4 *Theoretical Approximation of Adhesion Forces*

Adhesion Force of a Liquid Bridge.[6,7] The maximal tensile force that can be transmitted by a liquid bridge between two monosized spheres consists of two components:

(1) An adhesion force component A_c caused by the negative capillary pressure in the bridge:

$$A_c = p_c \cdot \frac{\pi}{4} \cdot x^2 \cdot \sin^2 \beta$$

(2) An adhesion force component A_b caused by the boundary force at the contact line solid–liquid–gaseous, which is determined by the surface tension of the liquid, α:

$$A_b = \alpha \cdot x \cdot \pi \cdot \sin \beta \cdot \sin(\beta + \delta)$$

By adding the two parallel adhesion force components A_c and A_b and after introducing the dimensionless function F_A a formula for the effective adhesion force A_L of a liquid bridge is obtained:

$$A_L = \alpha \cdot x \cdot F_A = \alpha \cdot x \cdot f_1\left(\beta, \delta, \frac{a}{x}\right) \quad (6.7)$$

where α is the surface tension of the liquid, x the diameter of the spherical, monosized particles, β the angle according to Figure 6.4, and a the distance of the particle surfaces at the coordination point.

Therefore, the adhesion force of a liquid bridge is proportional to the surface tension α, the particle diameter x, and a function of the angle β, the angle of contact δ, and the dimensionless quotient a/x. β defines the size of the liquid bridge and can be substituted by ϕ, the liquid volume divided by the volume of the solid particles:

$$\phi = \frac{V_b}{2 \cdot \pi \cdot x^3/6} = \frac{3}{\pi} \cdot f_2\left(\beta, \delta, \frac{a}{x}\right)$$

with V_b the volume of the liquid bridge.

By inserting the adhesion force A_L [Eq. (6.7)] into the basic formula, Eq. (6.4), and assuming that $k \cdot \epsilon \approx \pi$,[8] the maximal tensile strength σ_{tb} of agglomerates with liquid bridges becomes:

$$\delta_{tb} = \frac{1 - \epsilon}{\epsilon} \cdot \frac{\alpha}{x} \cdot F_A \quad (6.8)$$

Adhesion due to van der Waals Forces. Depending on the geometrical model (Fig. 6.5) being used and on the theoretical approach taken, different relationships exist for the approximation of adhesion by van der Waals forces. The best-known equations are those developed by Hamaker[9] based on the microscopic theory of London–Heitler. For the model sphere/plane (Fig. 6.5a), a distance $a < 100$ nm, and a particle diameter x, the adhesion force A_{v_1} is:

$$A_{v_1} = \frac{H}{12 \cdot a^2} \cdot x \quad (6.9)$$

For the model sphere/sphere (Fig. 6.5b), and the same limitations as mentioned above, Hamaker calculates an adhesion force A_{v_2}:

$$A_{v_2} = \frac{H}{24 \cdot a^2} \cdot x \quad (6.10)$$

H, the "Hamaker Constant," which depends on the material characteristics, has values in the order of 10^{-20} to 10^{-19} J.

More recently, Krupp[10] developed a formula for the model sphere/plane (Fig. 6.5a) which is based on the macroscopic calculations of Lifshitz–Landau:

$$A'_{v_1} = \frac{\hbar \bar{\omega}}{16 \cdot \pi \cdot a^2} \cdot x \quad (6.11)$$

$\hbar\bar{\omega}$ is the "Lifshitz–van der Waals Constant," which, depending on the material characteristics, has values between 0.2 and 9 eV (1 eV = $1.6 \cdot 10^{-19}$ J). All equations for the approximation of van der Waals forces differ only in the constants. The adhesion force A_v is always proportional to the particle diameter x and inversely proportional to the squared distance a:

$$A_v = c \cdot \frac{x}{a^2} \quad (6.12)$$

By inserting Eq. (6.12) into the basic formula, Eq. (6.4), and assuming that $k \cdot \epsilon \approx \pi$,[8] the maximal tensile strength σ_{tv} of agglomerates bound by van der Waals forces becomes:

$$\sigma_{tv} = \frac{1 - \epsilon}{\epsilon} \cdot \frac{c}{a^2} \cdot \frac{1}{x} \quad (6.13)$$

Adhesion due to Electrostatic Forces. In the case of electrostatic forces, one must distinguish between an excess charge and the electrical double layer (equilibrium).

The strength due to excess charges can be estimated if it is assumed that positively and negatively charged particles are arranged in a uniform pattern.[4] The basis for the derivation is Coulomb's formula for the attraction force between two spherical, nonconducting particles of equal size, the distance between which is much smaller than their diameter. If the charges $Q = \gamma \cdot \pi \cdot x^2$ are uniformly distributed on the surfaces the adhesion force A_c can be approximated by:

$$A_c = \frac{10\gamma^2}{(1 + a/x)^2} \cdot x^2 \quad (6.14)$$

For quartz the maximal charge density per unit area, γ, was estimated to be $\gamma^2_{max} \approx 0.25$ N/m^2.[1] If it is assumed that the charged particles forming an agglomerate are arranged like an ion lattice, then the attraction force between two adjacent, oppositely charged particles is approximately a factor 0.3 smaller because of the repulsion of neighboring particles with the same charge.

By inserting Eq. (6.14) into the basic formula, Eq. (6.4), and assuming that $k \cdot \epsilon \approx \pi$,[8] the maximal tensile strength σ_{tc} of agglomerated due to excess charges is:

$$\sigma_{tc} = \frac{1 - \epsilon}{\epsilon} \cdot \frac{3\gamma^2}{(1 + a/x)^2} \quad (6.15)$$

Because of the field character of this binding mechanism, the tensile strength is independent of the particle size. Also, the strength due to excess charges is very small, and the charges tend to equalize with time. Therefore, this mechanism is most often important only for the initial formation of agglomerates.

Much more important, however, are adhesion forces due to electrical double layers. This phenomenon can develop if the particles touch each other and is permanent. According to Krupp[10] the "attraction pressure" due to electrical double layers between two semi-infinite bodies is in the order of P_{el} 10^4 to 10^7 N/m^2 (10^5 to 10^8 dyn/cm^2). In comparison, the van der Waals attraction pressure between two semi-infinite bodies is P_{vdW} 2×10^7 to 3×10^8 N/m^2 (2×10^8 to 3×10^9 dyn/cm^2).

It may seem as if the two mechanisms exclude each other. However, since P_{vdW} decreases with $1/a^3$ and P_{el} stays almost constant even over macroscopic distance, the electrical double layer will contribute to the adhesion of particles, particularly if the contact surfaces are rough.

A theoretical approximation for specific systems is still not yet possible, since little is known about the distribution of charges in different materials. The effect of magnetic particles in agglomerates corresponds to that of excess charges and is subject to the same limitations.

6.2.3 Experimental Determination of Agglomerate Bonding and Strength

The most important techniques for the experimental determination of agglomerate strength known today measure crushing, shear, and tensile strengths. Sketches A–F in Figure 6.6 show schematically the methods for measuring

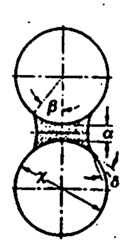


Figure 6.4. Schematic representation of a liquid bridge between two spherical, monosized particles.

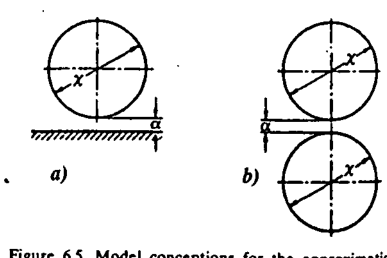


Figure 6.5. Model conceptions for the approximation of van der Waals adhesion. (a) Sphere/plane; (b)

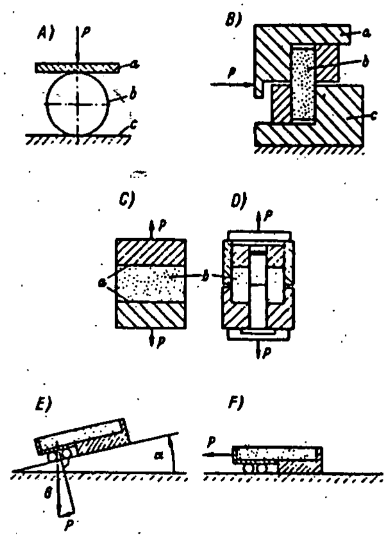


Figure 6.6. Methods for measuring the strength of agglomerates and particle conglomerates. (*A*) Determination of crushing strength: (*a*) loaded plate, (*b*) agglomerate, (*c*) support plate. (*B*) Determination of shear strength: (*a*) upper receptable, (*b*) compact or briquette, (*c*) lower receptacle. (*C*) and (*D*) Determination of tensile strength of strong agglomerates: (*a*) adhesive, (*b*) agglomerate (eventually machined). (*E*) and (*F*) Determination of tensile strength of weak agglomerates and of particle conglomerates.

the strength of agglomerates and particle conglomerates.

Figure 6.6A shows the determination of the crushing strength.[11] This method is a very simple one. Individual agglomerates are placed between two parallel plates and loaded with a uniformly increasing force P until failure occurs. Usually the "agglomerate strength" is defined as the mean statistical force at failure of a larger number of agglomerates tested by this method. Sometimes a crushing strength is calculated by dividing the force at failure by the projection area of the agglomerate; however, from a physical point of view this is not

The results of this test method are very rarely comparable. For spherical pellets the stressing is uniform only from test to test if all agglomerates are absolutely globular. In the case of agglomerates with flat ends the transverse expansion is blocked by friction between pellet and plate; thereby uncontrolled stress concentrations build up that can be the true cause for failure.

In Figure 6.6B, an apparatus is sketched for the investigation of shear strength. Originally this method was used in soil mechanics for the determination of shear curves of cohesive bulk solids. The "strength" of the conglomerate caused by internal friction can be determined graphically from the shear curves. The agglomerate must have two parallel surfaces, which may have to be produced by machining. The test specimen is fastened in the apparatus and stressed by the force P. The shear strength is defined by the shear force at failure divided by the shear plane.

Figure 6.6C shows in principle the "adhesive" method for the determination of tensile strength. Cylindrical agglomerates with two parallel and flat ends are centrically cemented between two so-called adaptors. To eliminate bending stresses it is necessary to machine spherical or nonsymmetrical agglomerates into cylindrical specimens using a special method (Fig. 6.7).[12] This sample is fastened to two thin wires (Fig. 6.8) and subjected to tensile forces in a conventional testing machine (Fig. 6.9a and b). The tensile strength of the agglomerate is defined as the tensile force P at failure divided by the cross section of the cylindrical specimen.

Figure 6.6D sketches the determination of the tensile strength of model agglomerates by means of the wall friction method.[7] For this test a cylindrical pellet—potentially with a central pin—is produced in a press. After removing the specimen from the press, it is stressed directly in the die shell. The tensile force is transmitted by adhesion between the end surfaces and the "pistons" as well as on the circumference and the die walls. Again,

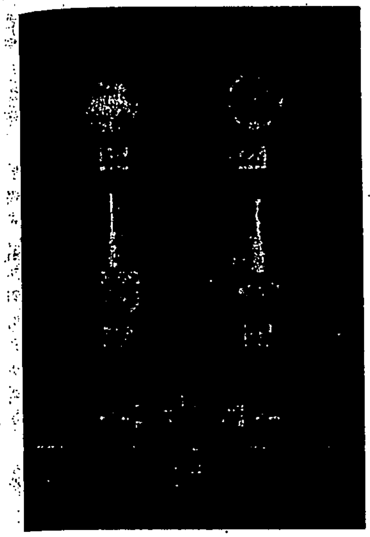


Figure 6.7. Preparation of "spherical" agglomerates for the determination of tensile strength by the "adhesive" method.[12]

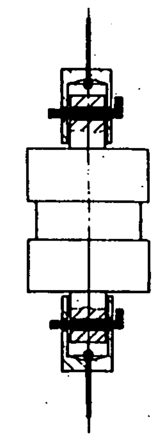


Figure 6.8. Schematic representation of the fastening method with thin wires.

rupture force P divided by the cross-section of the cylindrical or ring-shaped sample.

Figure 6.6E shows a method that is particularly suitable for the determination of low conglomerate strengths. The rupture stress is measured in a model arrangement. The powder to be investigated is filled into a flat, often rectangular split mold and densified by vibration or compaction. The upper half of the mold is fastened to a tiltable plate while the lower half is supported frictionless by rolls or spheres on the same plate. For the determination of the rupture force, the plate is slowly lifted at the end carrying the fixed mold until the particle conglomerate separates. The strength is defined as the force at failure divided by the cross-section of the agglomerate.

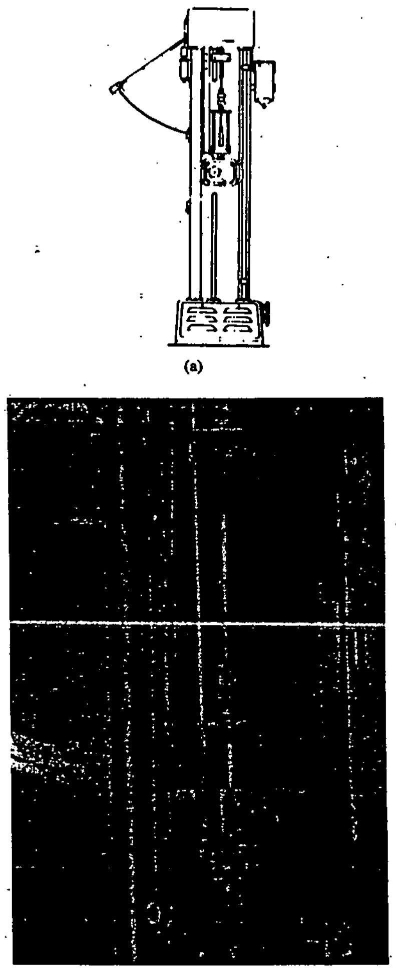
Another method based on the same principle uses a slowly increasing horizontal force P to pull apart the specimen (Fig. 6.6F) In this ing the entire test. The strength is defined as described above. Figure 6.10 shows a recent design of this experimental apparatus.[7] It is constructed such that two tests can be carried out on the same sample, and, if moist agglomerates are being investigated, the capillary pressure p_c can be measured simultaneously by means of a U-tube manometer. By means of inductive displacement gauges the expansion prior to rupture can be also determined.

6.2.4 Results of the Determination of Agglomerate Strength

6.2.4.1 Theoretical Approximation of Agglomerate Strength

[redacted] They often depend on the size of the particles forming the agglomerate and predict the maximum strength of conglomerates caused by a particular binding mechanism. Figure 6.11 shows the tensile strength σ_t versus particle size x in a double logarithmic plot.

The horizontal dotted line divides the entire



(a)

(b)

Figure 6.9. Testing machines adopted for the determination of the tensile strength of agglomerates. (*a*) Schematic overall view, (*b*) close-up during an actual tensile test.

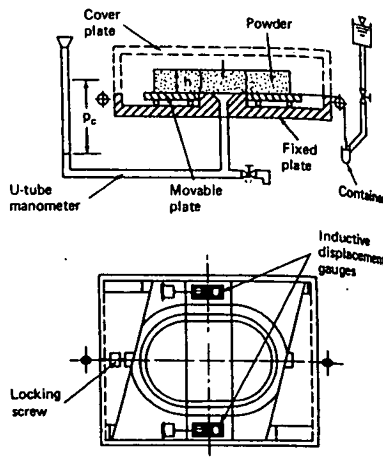


Figure 6.10. Diagrammatic representation of the split plate apparatus, according to Schubert.[13]

sent binding mechanisms that are independent of the size of the particles to be agglomerated. Region I describes high-pressure agglomeration. This technology uses high compaction pressures causing brittle disintegration as well as deformation of particles and favoring inter-

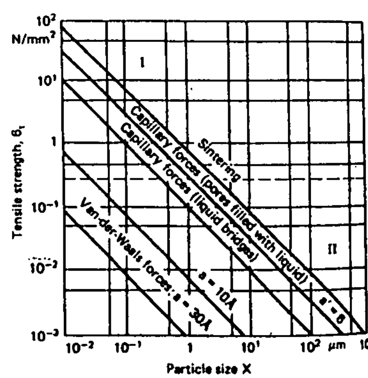


Figure 6.11. Approximation of the maximal theoretical tensile strength of agglomerates. Porosity: $\varepsilon = 0.35$. Region I: for example, hardening binders. Region II: Crystallizing soluble substances, for example, salts.

particle contact. Agglomerates with highly viscous and hardening binders are also included in this region. The lower region II describes the much weaker bonds caused by recrystallization of dissolved substances.

The diagonal lines define maximal tensile strengths, which depend on the size of the particles to be agglomerated. The van der Waals lines were calculated using the model sphere/sphere and particle distances of $a = 3$ and 1 nm. Assuming a distance of $a = 0.4$ nm (equilibrium distance) and using the model sphere/plane, the line would be pushed higher, close to the one representing capillary forces. If, in addition, plastic deformation of particles is considered, still higher agglomerate strengths can be obtained.

A narrow region characterizes the effect of liquid bridges. Somewhat higher is the line for the strength of agglomerates that are completely filled with a liquid. For this diagram it was calculated assuming water and the constant $a' = 6$. The strength of agglomerates with sinter and adhesion bridges can be expected above this line.

In each case, the predictions of Figure 6.11 are valid only for certain assumptions. In the following a few examples shall demonstrate the variability of the correlations if individual parameters are changed.

Figure 6.12a and b show salt bridges that were obtained at different drying temperatures[14] during a model experiment. The visual examination indicates that the drying temperature must play an important role in the development of agglomerate strength even if all other parameters are kept constant.

Capillary pressure and tensile strength of moist agglomerates are associated with each other. To a great degree they are influenced by the amount of liquid that is present in the pore volume of the agglomerate. Assuming that the liquid wets the solid particles ($\delta = 0$), a classification as shown in Figure 6.13 can be defined. It is valid for three-phase systems consisting of a disperse solid material and two

Figure 6.12. Common salt (NaCl) crystallizing between glass spheres (model experiment). (*a*) Drying at room temperature, (*b*) drying at 110°C.

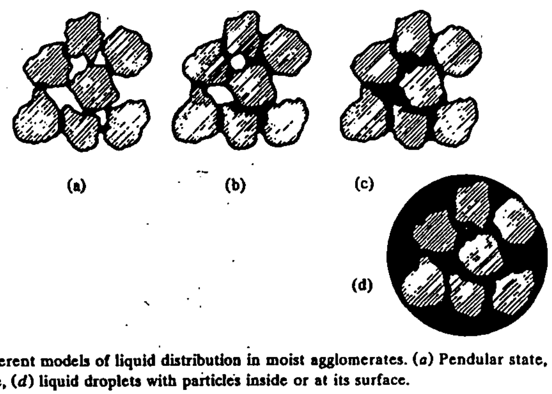


Figure 6.13. Different models of liquid distribution in moist agglomerates. (*a*) Pendular state, (*b*) funicular state, (*c*) capillary state, (*d*) liquid droplets with particles inside or at its surface.

immiscible fluid phases. The dark colored area represents the wetting fluid phase.

A small quantity of liquid causes liquid bridges between the particles forming the agglomerate (Fig. 6.13a). This region is called the pendular state. By increasing the amount of liquid, the funicular state is obtained (Fig. 6.13b) where both liquid bridges and pores filled with liquid are present. The capillary state (Fig. 6.13c) is reached when all pores are completely filled with the liquid, and concave menisci develop at the surface of the agglomerate. The last state (Fig. 6.13d), a liquid droplet with particles inside or at its surface, is an important mechanism for wet scrubbing and has relevance for agglomerate strength in spray dryer/agglomerators. Corresponding to the two patterns, Figures 6.13a and c, different models exist for the theoretical determination of agglomerate strength with a transition range in between (Fig. 6.13b).

Formerly, mathematical approximations were used for estimating the adhesion forces that can be transmitted through a liquid bridge. More recently, Schubert[15,16] developed exact equations for all rotationally symmetric liquid bridges. In Figure 6.14 the nondimensional force $F_A = A_L/\alpha \cdot x$ [Eq. (6.7)] is plotted versus V_b/V_s for various geometric situations, where V_b is the bridge volume, V_s the volume

spheres, α the surface tension of the liquid, the wetting angle $\delta = 0$, and a the distance at the coordination point. As the value of V_b/V_s increases, the attraction forces increase for planes and cones and decrease for spheres.

Under normal atmospheric conditions and with wetting solids it must be expected that liquid bridges are developing by a capillary condensation at contact points ($a = 0$). Depending on the contact geometry involved, the attractive fores resulting from this mechanism

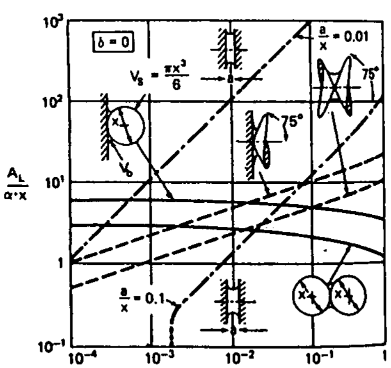


Figure 6.14. Computed adhesion forces resulting from

may exceed the van der Waals forces. For example, Figure 6.15 shows the nondimensional force $F_A = A_L/\alpha \cdot x$ [Eq. (6.7) as a function of the dimensionless distance at the coordination point a/x for the model sphere/sphere.[17] At $a/x = 0$ and $\beta \to 0$, the maximum value $A_L = \pi \cdot \alpha \cdot x$ is obtained. If the bridge is stretched the attraction force decreases the more, the smaller the liquid volume.

Assuming complete wetting ($\sigma = 0$), the correlation between the nondimensional force $F_A = A_L/\alpha \cdot x$ [Eq. (6.7)] and the filling angle β (Fig. 6.4) at different values of a/x is presented in Figure 6.16. For spheres in contact ($a/x = 0$) the value of $F_A = A_L/\alpha \cdot x$ decreases from π (at $\beta = 0$) as the liquid content increases. For finite values of a/x, however, the curves pass through a maximum or, respectively, increase with higher liquid saturation.

The dotted curve in Figure 6.16 divides the graph into a field of capillary excess pressure ($F_{P_c} < 0$) and a field of capillary suction ($F_{P_c} > 0$). For comparison, two curves were calculated for $a/x = 0.1$, one representing the exact theory of Schubert[15,16] and the other an earlier approximation by Pietsch and Rumpf.[6]

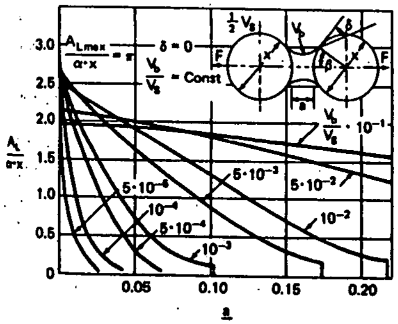


Figure 6.15. Nondimensional adhesion force $F_A = A_L/\alpha \cdot x$ as a function of the dimensionless distance at the coordination point a/x.[17]

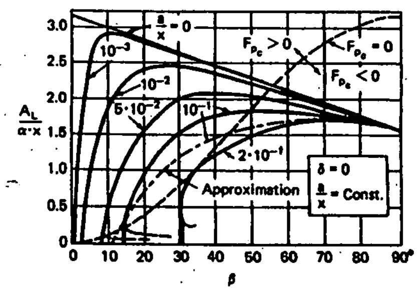


Figure 6.16. Correlation between the dimensionless adhesion force $F_A = A_L/\alpha \cdot x$ between two equal spheres and the filling angle β. Assumptions: $\delta = 0$ and a/x = constant.

For the determination of the maximally transferable uniaxial tensile stress Eq. (6.8) can be rewritten:

$$\frac{\sigma_{tb} \cdot x}{\alpha} = \frac{1-\epsilon}{\epsilon} \cdot F_A'(\beta, \delta, a/x) \quad (6.16)$$

For the transition range (Fig. 6.13b), the easily measurable liquid saturation S is introduced:

$$S = \pi \cdot \frac{1-\epsilon}{\epsilon} \cdot \phi \quad (6.17)$$

It is defined as the ratio of liquid volume to pore volume of the agglomerate. Thus, Eq. (6.16) yields:

$$\frac{\sigma_{tb} \cdot x}{\alpha} = \frac{1-\epsilon}{\epsilon} \cdot F_A''(\epsilon, S, \delta, a/x) \quad (6.18)$$

Equation (6.18) can be used to compute the correlation between the value $\sigma_{tb} \cdot x/\alpha$ and the liquid saturation S. Results for two different porosities ϵ and different distance ratios a/x (assuming complete wetting [$\delta = 0$]) are presented in Figure 6.17. Again, for comparison, the curves for $\epsilon = 0.35$ and $a/x = 0.1$ representing the exact theory and the approximation, respectively, have been included in Figure 6.17.

In the capillary state the strength of the agglomerate is determined by the capillary suction. Since only those pores filled with the

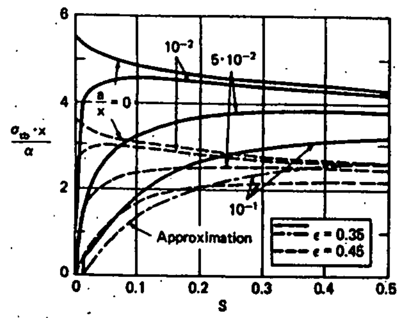


Figure 6.17. Maximally transferable uniaxial tensile stress $\sigma_{tb} \cdot x/\alpha$ as a function of the liquid saturation S of moist agglomerates. Assumption: complete wetting ($\delta = 0$).

liquid contribute to the strength, Eq. (6.1) must be rewritten:

$$\sigma_{tc} = S \cdot p_c \qquad (6.19)$$

Equation (6.19) assumes that the liquid is uniformly distributed in the agglomerate. The product $S \cdot p_c$ can be calculated from the capillary pressure/saturation curve (Fig. 6.18b). By definition, the starting point of capillary pressure curves is $p_c = 0$ at $S = 1$. During drainage of the agglomerate the capillary pressure follows the curve marked in Figure 6.18b until it reaches a point at which only isolated capillaries exist. If, starting at that point, liquid is reintroduced into the agglomerate, $p_c = 0$ is reached at $S < 1$, as not all pores can be filled with liquid by imbibition. The remaining air pockets are blocked off by adjacent pores that are already filled. Repeated drainage/imbibition tests lead to the typical hysteresis loop shown in Figure 6.18b. It is explained by the existence of pore bulges and pore necks as well as by the contact angle hysteresis.[18]

Figure 6.18a shows schematically the maximally transferable tensile stress σ_t as a function of the liquid saturation S. The capillary pressure curve (Fig. 6.18b) is used to calculate $\sigma_{tc} = S \cdot p_c$ for $S_c \leq S \leq 1$. The capillary state ends when liquid bridges between the particles start forming ($S \leq S_c$). The funicular state exists in the region $S_b \leq S \leq S_c$. For the pendular state ($S \leq S_b$) the already discussed Eq. (6.18) is valid.

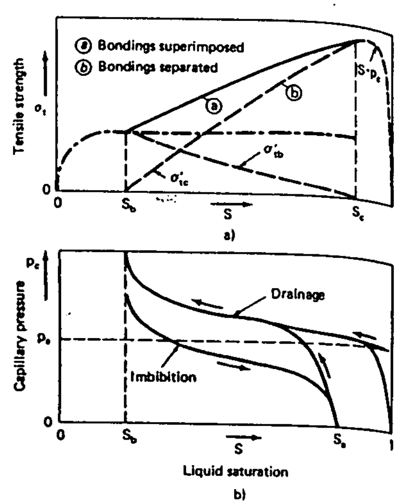


Figure 6.18. (*a*) Maximally transferable tensile stress σ_t and (*b*) capillary pressure p_c as a function of the liquid saturation S.

In the transition range, the funicular state ($S_b \leq S \leq S_c$), in which liquid bridges coexist with liquid-filled pores, two cases can be constructed which follow a model published first by Rumpf.[1]

1. Both bonding mechanisms can be superimposed.
2. Each of the bonding mechanisms acts alone.

Assuming that the ratio of the liquid in the bridges to the total liquid diminishes linearly from 1 at S_b to 0 at S_c, one obtains the following for the individual bonding mechanisms (see Fig. 6.18a):

$$\sigma'_{tb} = \sigma_{tb}(S_c - S)/(S_c - S_b) \qquad (6.20)$$

$$\sigma'_{tc} = p_c \cdot S_c(S - S_b)/(S_c - S_b) \qquad (6.21)$$

If both mechanisms act alone curve *b* in Figure 6.18a represents the expected results. If bonding mechanisms can be superimposed curve *a* results from the sum σ'_{tb} plus σ'_{tc}.

In all those cases where adhesion is caused by van der Waals or electrostatic attraction or by liquid bridges, surface roughness reduces the maximally transferable adhesion force. For the model sphere/plane and van der Waals attraction, Figure 6.19 shows the controlling radii for the calculation of the adhesion force according to Eq. (6.11). The shape of particles with surface roughness can be approximated by superimposing two spheres. The large radius R is considered the equivalent radius of a sphere of same volume as the particle, whereas the small radius r represents the surface roughness.[17]

Considering the model of Figure 6.19 and Eq. (6.11), the highest attraction forces A_{max} must be obtained if the adhesion partners are in contact and have smooth surfaces. Contrary to the indication of Eq. (6.11), the attraction force on contact is in reality finite. Therefore, an adjustment parameter Z_0 must be introduced:

$$A''_{v_1} = \frac{\hbar\bar{\omega}}{8\pi(a + Z_0)^2} \cdot R \qquad (6.22)$$

Krupp[10] has defined $Z_0 = 4 \cdot 10^{-8}$ cm (0.4 nm) as a measure for the atomic distance. For Figure 6.20b, A_{max} was calculated for different adhesion mechanisms using this value of Z_0, although it does not represent a true atomic distance. Rather, it is an approximate or adaptive parameter, the accurate value of which needs still to be determined.

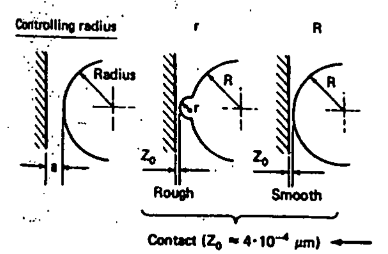


Figure 6.19. van der Waals model sphere/plate with and without surface roughness.[17]

Figure 6.20b shows for the model sphere/plate the correlation between the maximum adhesion force A_{max} on contact and the diameter $x = 2R$ of smooth particles for different adhesion mechanisms. The highest attraction forces are caused by liquid bridges assuming complete wetting ($\delta = 0$) and water as the liquid. Van der Waals forces are smaller by almost an order of magnitude, although a relatively high Lifshitz–van der Waals constant ($\hbar\bar{\omega} = 5$ eV $= 8 \cdot 10^{-19}$ J) was chosen. If two different materials contact, an electrostatic attraction force develops that is caused by the contact potential. The latter depends on the characteristics of the two contacting materials and their surface conditions. Again, the potential chosen ($U = 0.5$ V) represents a relatively high value. For conductors the electrostatic attraction force is higher than for nonconductors with the same contact potential because the charge is concentrated at the surface. Electrostatic attraction forces can also result from excess charges originating from friction, crushing, or electron and, respectively, ion adsorption. The highest possible excess charges are around 10^2 elementary charges $e/\mu m^2$.

Figure 6.20b indicates that for smooth spheres with sizes below 100 μm the electrostatic adhesion is negligible compared with van der Waals forces and even more so in relation to forces caused by liquid bridges.

Figure 6.20a describes the influence of surface roughness, represented by r (abscissa), on the attraction force A for different adhesion mechanisms. The curves were calculated for spheres with constant diameter $x = 2R = 10$ μm. The corresponding values of A_{max} can be determined in Figure 6.20b. Only for van der Waals forces two further curves for $R = 0.5$ μm and $R = 50$ μm were plotted since—because of their short-range character—the influence of roughness on van der Waals forces is very pronounced.

Investigating the curve for $R = 5$ μm and van der Waals attraction, the following observations can be made. At $r = 0$ (not shown in

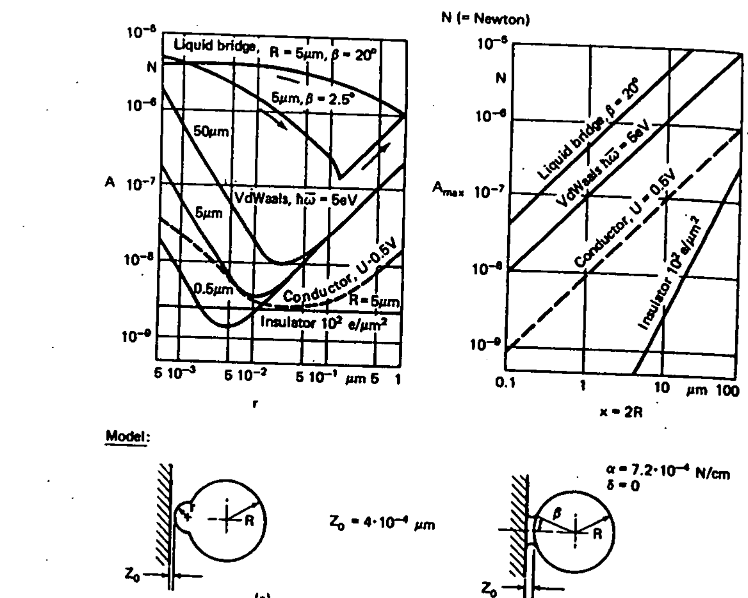


Figure 6.20. Attraction forces caused by different adhesion mechanisms for the model sphere/plate. Contact $a = 0$, $Z_0 = 0.4$ nm.[17] (*a*) Influence of the roughness radius r on the attraction force A. (*b*) Influence of the diameter R of the smooth spheres on the attraction force A_{max}.

Fig. 6.20a) the maximum adhesion force $A = A_{max} = 10^{-6}$ N is obtained (Fig. 6.20b). With increasing r, the distance of the larger sphere from the plane grows, and the adhesion force decreases proportional to r^{-2}. Later the influence of the large sphere diminishes, and a minimum is reached at which both attraction forces act simultaneously. A_{min} is only approximately $1/250\ A_{max}$. If r grows further, the influence of the large sphere disappears. Then, only the attraction force of the small sphere remains, which increases proportional to r. With growing R, r_{min} and the corresponding A_{min} increase but not at the same rate as A_{max}. At $R = 50\ \mu m$, A_{min}/A_{max} equals $1/1000$.

Liquid bridges are much less sensitive to surface roughness. If the angle β is not too small, for example, $\beta = 20°$, the roughness is immersed in the liquid and merely increases the distance a. The smaller β, the more pronounced is this influence. At $\beta = 2.5°$ the liquid bridge breaks off from the large sphere at $\sim r = 10^{-1}\ \mu m$ and remains on the small sphere. Then, the attraction force increases proportional to r. $\beta = 2.5°$ can already correspond to capillary condensation. If β is still smaller, the transition to the line proportional to r occurs at smaller values of r, for example, for $\beta = 1°$ at $r \approx 5 \cdot 10^{-2}\ \mu m$. In any case, the attraction force due to capillary condensation at the roughness peaks is always larger than the van der Waals force.

The electrostatic attraction forces of electrical conductors and of insulators with excess charges are smaller than the van der Waals forces. However, the influence of roughness is less pronounced and disappears completely for

conducting particles facing a plane with an opposite charge of the same density.

For a long time the opinion existed that, compared with van der Waals forces electrostatic attraction is always negligible. Based on the knowledge of the independence of roughness on electrostatic adhesion, this must now be corrected. Since the electrostatic attraction force increases with R^2, the 50 μm sphere with a charge density of $10^2\ e/\mu m^2$ would generate a higher value that could be obtained with the van der Waals model in much of the range of roughness shown in figure 6.20a. Therefore, in dry agglomerates formed by relatively large particles the electrostatic attraction forces due to excess charges—which do not depend on surface roughness—may have an important share in agglomerate strength.

The following results are generally valid:

1. For adhesion, forces caused by liquid bridges are most important and, normally, represent the highest share. Even in dry systems—due to capillary condensation—liquid bridges may be the controlling mechanism.
2. Van der Waals forces are extremely sensitive to surface roughness but should always be larger than forces caused by the contact potential.
3. Excess charges can also be a controlling factor for adhesion, particularly if relatively large particles form a dry conglomerate.
4. At distances in excess of 1 μm or, respectively, $a/x > 0.2$, only electrostatic forces are effective. They cause particle attraction before adhesion takes over and forms agglomerates.

6.2.4.2 Results of Experimental Determinations of Agglomerate Strength

In the following, some results of experimental investigations will show that if suitable model materials, agglomeration techniques, and experimental stressing methods are used, the results can be compared to Sections 6.2.2 and 6.2.4 (see Theoretical Approximation of Agglomerate Strength) can be well approximated.

Crystallization of Dissolved Substances During Drying[14]. If drying of agglomerates that are filled with a salt solution starts at high liquid saturation S, evaporation begins at the surface of the agglomerate. The liquid flows to the surface by means of capillary suction. There, the dissolved substance crystallizes, forms a crust, and decisively controls further drying of the porous body. The crystal structure can therefore by influenced by either the drying temperature or the presence of a crust. In the latter case, the tensile strength of the agglomerate changes due to the varying drying conditions even if the strength is measured on the core after removing the crust.

A typical example is shown in Figure 6.21. The tensile strength, σ_t, is plotted versus the drying temperature, t_d. The parameter is the liquid saturation S. This diagram was obtained using a nearly saturated salt (sodium chloride) solution and a narrow limestone powder fraction. At very small liquid saturations ($S \leq 20\%$, curves *a* and *b*) no crust is built up, and the tensile strength increases with the drying temperature. This rise is caused first by the increasing crystallization velocity and second by the amount of salt forming bridges that changes almost proportionally to the liquid saturation. At a liquid saturation of 20% the formation of a thin crust starts to influence the tensile strength slightly. At the highest liquid saturations examined ($S \geq 45\%$, curves *e* and *f*), the dense crust is at all temperatures the deciding factor for drying and for the crystallization velocity. Their core tensile strength is low and remains almost constant. Such agglomerates burst at high drying temperatures much like a pressure vessel (Fig. 6.22). The high tensile strength of agglomerates with $S = 30\%$ dried at a temperature of 350°C (curve *d*) is caused by small hair fractures in the crust that did not cause the ag-

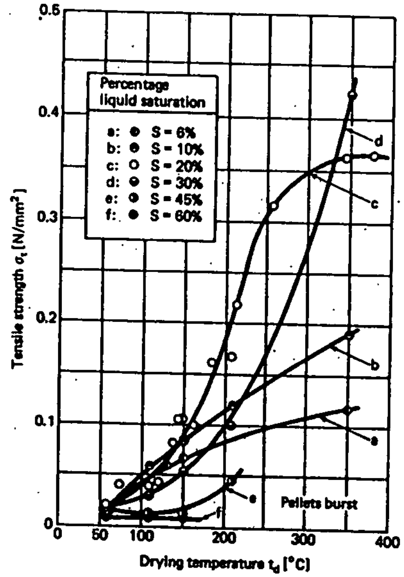


Figure 6.21. Tensile strength of σ_t of the core of agglomerates (crust removed) with salt bridges as a function of the drying temperature t_d at different liquid saturations S before drying.[14]

rate and thus increased the tensile strength of the dry agglomerate core.[19]

Figure 6.23 shows the same set of results as shown in Figure 6.21 but plotted in a different way. This time the tensile strength σ_t is presented as a function of the liquid saturation S before drying. The parameter is the drying temperature t_d. This graph confirms that normally the highest strength is obtained at $S =$ 20% if it is measured after removing the crust. However, an optimum drying temperature exists whereby an agglomerate dries quickly but does not build up enough inside pressure to cause cracking or disintegration. Then, the tensile strength can also be determined with crust.

Tests to investigate this optimum drying temperature were carried out in an instru-

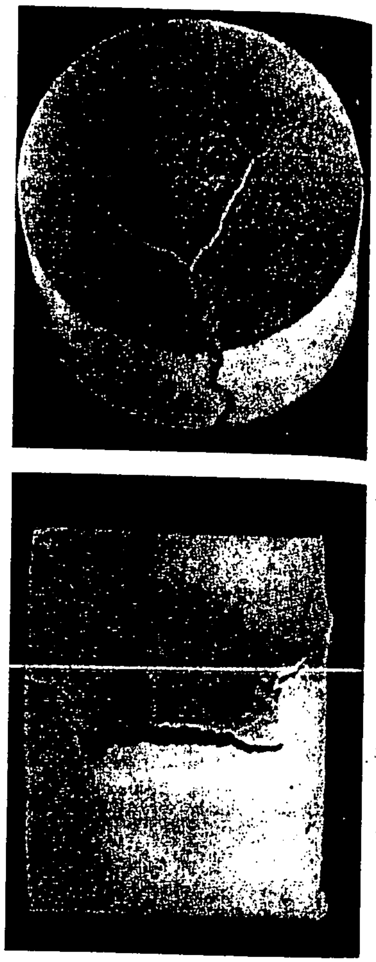


Figure 6.22. Cylindrical agglomerate that contained as NaCl solution and burst during drying.

mented drying channel.[20] Figure 6.24 shows a result. Below $S = 0.2$ the strength values for agglomerates with and without crust are identical. At saturations above 0.2 the strength of agglomerates with crust increases proportionally

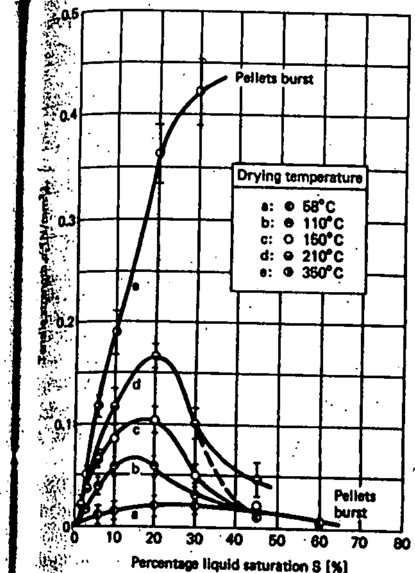


Figure 6.23. Tensile strength σ_t of the core of agglomerates (crust removed) with salt bridges as a function of the liquid saturation S before drying at different drying temperatures t_d.[14]

For practical applications the following conclusion can be drawn: To obtain high strength, drying should be carried out at the highest possible (without cracking) temperature using a saturated solution. Charé[20] found further that the air velocity does not substantially change the drying rate, and, therefore, the agglomerate strength.

[redacted]s. Agglomerates that are being built up by balling, that is the snowball-like forming of pellets in drums or discs, are nearly saturated with liquid. Figure 6.25 shows results of the determination of tensile strength plotted versus particle size. x_0 is the surface equivalent diameter and x_1 is the maximum of the diameter distribution. The diagonals represent the maximally tra...

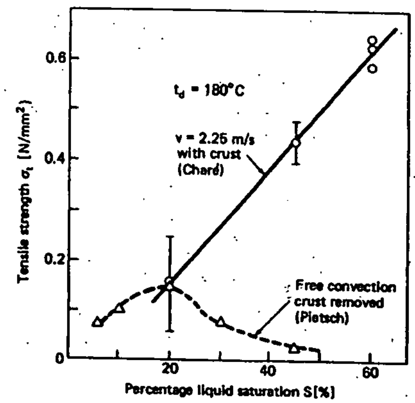


Figure 6.24. Tensile strength σ_t of agglomerates with salt bridges as a function of the liquid saturation S before drying with and without the crust.[21]

with Eq. (6.2) using $a' = 6$ and $a' = 8$. The diagram shows that $\sigma_t \sim 1/x$ is fulfilled. Values lower than theoretically predicted are mostly due to the fact that the agglomerates were not fully saturated when the tensile strength was determined.

The relationship $\sigma_t \sim \alpha$ was confirmed by Conway-Jones[11] with compression tests on spherical agglomerates (Fig. 6.26) and $\sigma_t x/\alpha \sim (1-\epsilon)/\epsilon$ was checked by Schubert,[15] who confirmed this correlation, too (Fig. 6.27).

It can be assumed that up to saturations of ≈20% to 40% the liquid in moist agglomerates is present in the form of discrete liquid bridges at the contact and coordination points between the particles forming the agglomerate. The tensile strength of such an agglomerate is predicted by Eq. (6.8). Experimentally it was investigated with the wall friction method (Figure 6.6D) using pellets made of narrowly distributed limestone powder and distilled water. In Figure 6.28 the experimental results are shown in comparison to the theory. The curves were approximated by varying the distance a (respectively, a/x). They seem to fit the exper-

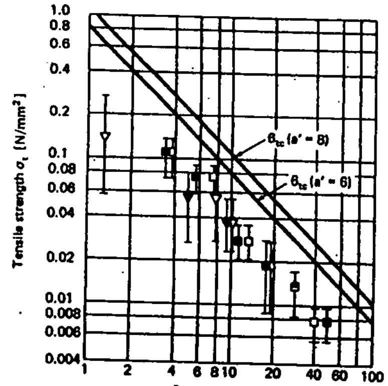


Figure 6.25. Tensile strength of moist agglomerates with high liquid saturation as a function of particle size. $\epsilon = 0.35$. Quartz powder: ▽: x_0, ▼: x_1. Limestone powder: □: x_0, ■: x_1.

a/x are purely empirical. However, considering the surface roughness of the particles the a/x values may be in the correct order of magnitude.

ALCOHOL/WATER MIXTURES

VOL. %		α	σ
ALCOHOL	WATER	(N/cm)	(N/mm²)
0	100	72.2×10^{-5}	8.31×10^{-1}
10	90	50.2×10^{-5}	6.02×10^{-1}
30	70	35.0×10^{-5}	4.53×10^{-1}
100	0	22.2×10^{-5}	2.42×10^{-1}

At higher liquid saturations more and more pores fill up and the models *liquid bridges* and *saturated pores* coexist. The theories described in Section 6.2.4 (Figs. 6.13, 6.17, and 6.18) where checked by Schubert.[15] Figure 6.29 shows the results. The tensile strength is plotted versus the liquid saturation and compared with the theory. At $S = 1$ the capillary pressure is zero. If, starting at this point, the agglomerate is drained, the capillary pressure raises steeply and then turns into a flatter curve. The point where the two tangents meet is defined as the so called entry suction pressure p_c. It is generally located near $S = 0.9$. Approximately at this point the maximum tensile strength of moist agglomerates exists. At lower and higher saturations the strength decreases. The results show that between $0.3 < S < 0.9$ both mechanisms contribute to the strength of agglomerates.

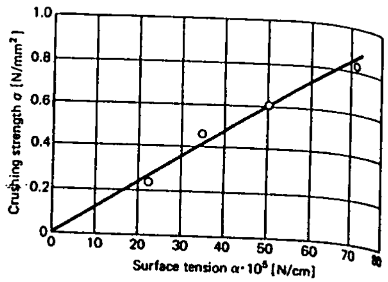


Figure 6.26. Relationship between crushing strength σ of moist agglomerates and surface tension α of the liquid.[11]

The capillary pressure and, therefore, the tensile strength are much larger if the liquid is drained than after imbibition. This knowledge can be very important for agglomeration and certain other technologies, for example, filtration (strength of filter cakes).

Strength due to van der Waals Forces

Because of the short range of van der Waals forces, particles forming an agglomerate

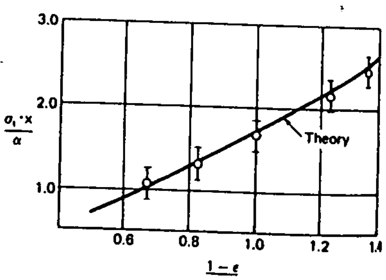


Figure 6.27. Relative tensile strength $\sigma_t \cdot x/\alpha$ of moist conglomerates formed of glass spheres plotted versus the porosity function $(1-\epsilon)/\epsilon$. Comparison between theory [Eq. (6.8)] and experiment.[15]

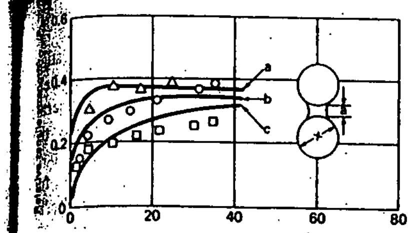


Figure 6.28. Relative tensile strength σ_t/p_c of agglomerates with liquid bridges as a function of the liquid saturation S. Limestone powder; △: $x_0 = 71\ \mu m$; ○: $x_0 = 35\ \mu m$; □: $x_0 = 13\ \mu m$.
a $\epsilon = 0.45$; $a/x = 0.02$ ($a = 1.4\ \mu m$)
b $\epsilon = 0.45$; $a/x = 0.04$ ($a = 1.4\ \mu m$)
c $\epsilon = 0.50$; $a/x = 0.1$ ($a = 1.3\ \mu m$)

[redacted] excludes other binding mechanisms.

The influence of adsorption layers on agglomerate strength was demonstrated using, respectively, air-dry material and powder, which was dehydrated at a temperature of 600°C prior to pressing it into pellets at 10^{-5} mbar and room temperature. Figure 6.30 shows the results[22]. [redacted] Pellets that were produced at atmospheric conditions from air-dry material (L) and, therefore, contain adsorbed water exhibit higher strength than those pressed at high vacuum from desorbed barite (HV). This is in general agreement with the expected influence of water adsorption on adhesion discussed above.

Herrmann[23] investigated in more detail the influence of water adsorption on the tensile and shear strength of barium sulfate briquets. Figure 6.31 shows some results. The tensile and shear strengths were determined on briquets produced and stressed in a high vacuum and at varying levels of relative humidity of the surrounding atmosphere. The normal relative humidity lies between 60% and 80%. Therefore, in the common sense, the powder must be considered dry. The strength is plotted in both parts of Figure 6.31 versus the relative water vapor pressure p/p_0 (with p_0 = water vapor pressure at saturation). The following conclusions can be drawn:

1. The tensile strength σ_t increases with growing relative water vapor pressure p/p_0. Responsible for this rise is the capillary condensation. Van der Waals forces participate only to a small extent.

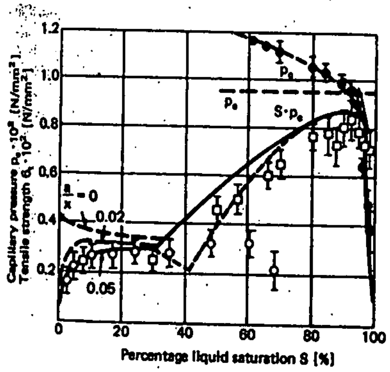


Figure 6.29. Tensile strength σ_t and capillary pressure p_c as a function of liquid saturation S. Limestone: $x_0 = 71\ \mu m$, $\epsilon = 0.415$. ●: p_c; □: σ_t drainage; ○: σ_t imbibition

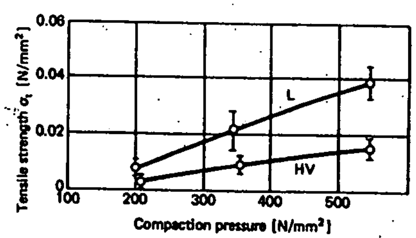


Figure 6.30. Tensile strength σ_t of barium sulfate pellets with (L) and without (HV) adsorption layers as a function of compaction pressure. Particle size of start-

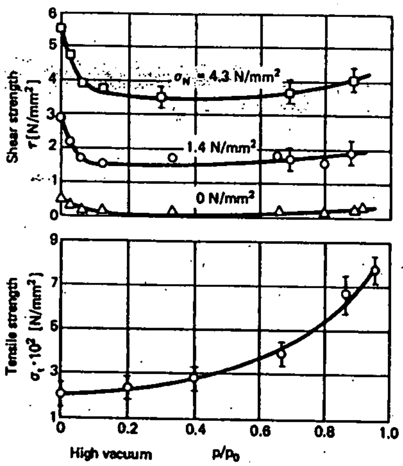


Figure 6.31. Shear strength τ and tensile strengths σ_t of barite briquettes as functions of the relative water vapor pressure p/p_0.[23] Particle size of starting material: 50 to 100 μm. Compaction pressures: 2.2×10^2 N/mm² (shear strength graph; 4.8×10^2 N/mm² (tensile strength graph).

2. Due to interparticle friction the shear strength τ is high in a high vacuum. This explains, for example, the extremely well-developed footprints that were visible during the first moon landing of man in the loose dust (Apollo II). Interparticle friction is highest at space conditions. With increasing relative humidity the shear strength τ first decreases rapidly. This is due to the fact that liquid films "lubricate" the particles and the friction decreases. Later, liquid bridges develop by capillary condensation and the strength increases again slowly.
3. At a normal load of $\sigma_N = 0$ the tensile strength σ_t of barite briquets is smaller than the shear strength τ. Therefore, the tensile strength is the critical strength, and failure occurs under tensile load.
4. The shear strength τ depends on the normal load σ_N which acts upon the specimen during shearing.

6.2.4.3 *Other Investigations*

A large number of other, specific investigations was carried out by various researchers, confirming still more theories of agglomerate bonding and strength. However, since this chapter is only meant to introduce some basic theoretical and experimental information, further results should be obtained from the respective scientific and technical literature.

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6.3 SIZE ENLARGEMENT BY AGGLOMERATION IN INDUSTRY

6.3.1 Parameters of Size Enlargement by Agglomeration

A basic equation for the tensile strength of agglomerates bonded by forces transmitted at the coordination points between particles was first developed by Rumpf.[1] It is [see also Eq. (6.4)]:

$$\sigma_t = \frac{1-\epsilon}{\pi} k \frac{\sum_{i=1}^{n} A_i(x, \ldots)}{x^2} \qquad (6.23)$$

where

σ_t = tensile strength
ϵ = porosity (= relative pore space)
$(1-\epsilon)$ = relative amount of solids
k = number of coordination points
[illegible]

With

$$k \cdot \epsilon \approx \pi \qquad (6.24)$$

and taking into consideration that most adhesion forces A_i are a function of the particle size x, Eq. (6.23) becomes:

$$\sigma_t = \frac{1-\epsilon}{\epsilon} \frac{\sum_{i=1}^{n} A_i(\cdots)}{x} \qquad (6.25)$$

Even after eliminating the influence of the particle size x the adhesion force A_i remains a function of several parameters that vary with the binding mechanism.

The particle size representing a distribution of irregularly shaped particles (= the real conditions) can be described for all models by the surface equivalent diameter x_0, the diameter of monosized spherical particles producing the same specific surface area (e.g., in m²/g) as the actual particle size distribution.

A similar equation describes the tensile strength of agglomerates which are held together by the negative capillary pressure of a liquid filling the pore space [see also Eq. (6.2)]:

$$\sigma_t = c \frac{1-\epsilon}{\epsilon} \alpha \frac{1}{x_0} \qquad (6.26)$$

where

c = constant between approx. 6 and 8
α = surface tension of liquid.

Equations (6.25) and (6.26) suggest that the strength of agglomerates is strongly influenced by the porosity and, respectively, the relative amount of solids and that it increases with the specific surface area (= decreasing surface equivalent diameter x_0) of the particulate matter forming the agglomerate. The latter also indicates that the presence or lack of very fine particles will favor or hinder the formation of strong agglomerates. In the case of wet agglomerates (capillary model) the surface tension participates directly in strength; it must be understood, however, that capillary forces provide only temporary bonding; post treatment will activate other binding mechanisms

Characteristics of agglomerates from particulate solids and a matrix binder, for example, cement in concrete-like aggregates, depend on the strength of all participating materials as well as their adhesion conditions and follow different relationships (see Section 6.2.2). Strong, highly impermeable, and leach-proof agglomerates are obtained if the distribution of the particulate matter favors the formation of dense structures as shown by the model in Figure 6.32.

[redacted]

Th[redacted] Equations (6.25) and (6.26)] [redacted]. Since, additionally, the representative particle size that best describes the phenomenon of agglomeration is the surface equivalent diameter x_0, the tendency of particulate matter to agglomerate increases greatly with the presence of very fine particulate matter.

The strong influence of small particles on x_0 can be easily demonstrated by the fact that a single spherical particle with density 1 g/cm³ having a mass of 1 g (particle diameter approx. 12.4 mm) features a surface area of approx. 4.8×10^{-4} m²/g. If this mass of 1 g is made up by (1.9×10^{12}) monosized 1 μm spherical particles of the same material and with porosity 0.4 the specific surface area is approx. 9.3 m²/g or more than four orders of magnitude larger (Table 6.4).

When determining the specific surface, methods must be chosen that measure only the outer particle surface and exclude accessible inner surface due to open particle porosity. Good estimates for this type of specific surface area of particulate matter can be obtained by the simple Blaine (permeability) method or the scanning of particle images.

Figure 6.33 is a dispersity scale indicating the most likely ranges for certain particulate matter (with particular reference to fines occurring in environmental control technologies) and the general applicability of agglomeration methods. Of course, the individual ranges do

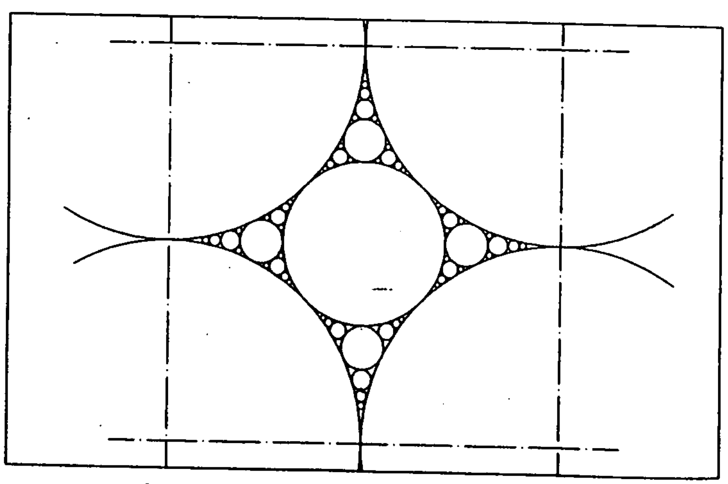


Figure 6.32. Model (cubic) depicting dense packing structure

...4. Some Characteristics of Spherical ...s (Density of Solid: 1 g/cm³).

VOLUME (cm³)	DIAMETER (mm)	NUMBER (—)	SURFACE AREA (m²/g)
1	12.4	1	$4.8\ 10^{-4}$
1.67	10^{-3}	$1.9 \cdot 10^{12}$	9.3

...ap and can be influenced by special con...s or processes.

...a particle size range below approx. 10 ...the natural attraction forces, such as ...ular (van der Waals), magnetic, and ...rostatic forces, which may be enhanced by ...ption layers, liquid films, or "binder" ...icals (e.g., flocculation agents) become ...ificantly larger than the separating forces ...to particle mass and external influences (...drag and centrifugal forces) so that adhe...occurs (Fig. 6.34). Because the probability of particle-to-particle collisions, which are pre...ditions for adhesion, rises with concentra...tion, the tendency of particulate matter to naturally agglomerate increases (for example, in a fluidized bed environment) but, independent of concentration, decreases with particle size despite their greater adhesion potential. The latter is due to the fact that ultrafine particles tend to follow flow lines so that collisions do not occur as frequently.

The natural agglomeration of "submicron" particles is a reason for the relatively high efficiency of many pollution control devices that separate such solids from process effluents. The effect can be increased by forcing the particles into increased motion, for example, in the case of smokes by the application of sound.

6.3.2.1 *Undesired Agglomeration*

Knowing the possible binding mechanisms of agglomeration and that, with few exceptions, bonding and strength of agglomerates is strongly influenced by particle size or surface, the reasons for and potential methods for the prevention of unwanted agglomeration phenomena during processing, storage, and han-

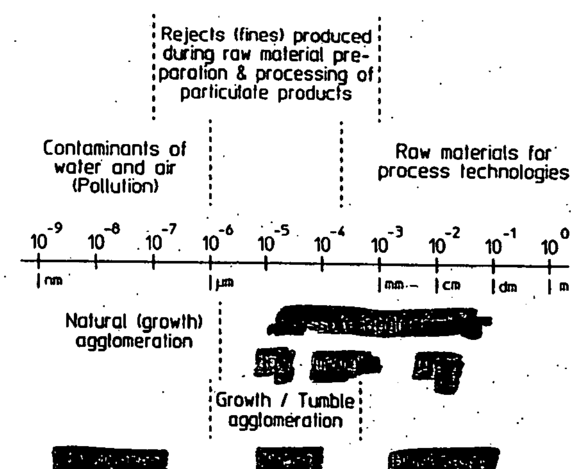


Figure 6.33 Dispersity scale ...

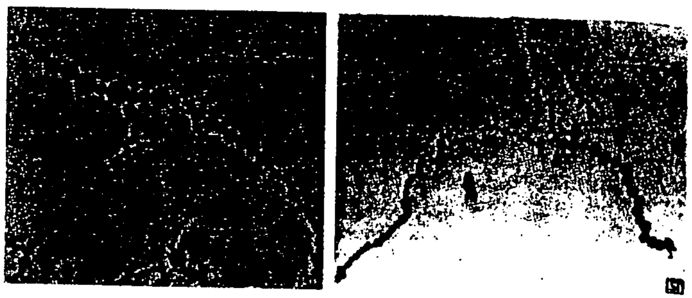


Figure 6.34. Chain-like natural agglomerates of "brown smoke" from steel converters formed by the combined effects of magnetic, electrostatic, and molecular attraction.[2]

dling of particulate solids are comprehensible. In most cases undesired agglomeration phenomena begin with the finer portion of the particle mass. In the following some examples[2] will be presented.

Comminution. During fine grinding in tube or roller mills deposits begin to form at a certain fineness, in the case of all materials, whereby two types of phenomena can be distinguished.

In the first case, the finest particles start to adhere to walls of grinding media in the mill, forming thin layers. On this basis coarser particles find excellent conditions for adhesion and massive deposits form rapidly. Experiments by Ocepek,[3] who investigated the particle size distribution across thick layers of build-up, showed that the finest particles are indeed found in the lowest layers. Figure 6.35 shows grinding balls which, after a short period of operation, are already covered with a light primary deposit, upon which additional layers will build up during extended grinding. Figure 6.36 is the photograph of the manhole cover of a ball mill, illustrating the extent of such deposits. These adhering layers produce a cushioning effect which lowers the intensity of stressing and, therefore, increases the duration of grinding.

The second phenomenon during dry fine grinding is the occurrence of agglomerates in the freely moving charge itself. Again, such agglomerates form only in the presence of a sufficiently large amount of fine particles and are frequently lamellar.

Agglomeration and adhesion in mills can be attributed to various bonding mechanisms. Since the mill housing often becomes highly charged by friction between its contents and the walls, electrostatic forces are often the cause of build-ups. This effect can be eliminated quite easily by grounding the mill. In other cases, wall deposits will begin with particles of a size that generally corresponds to that of the wall roughness. The strength of the deposited layer depends on the intensity of contact pressure which is magnified by the mill charge consisting of grinding media and mate-

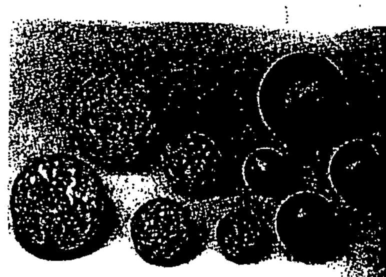


Figure 6.35. Grinding balls before (*right*) and after brief grinding (*left*).[2]

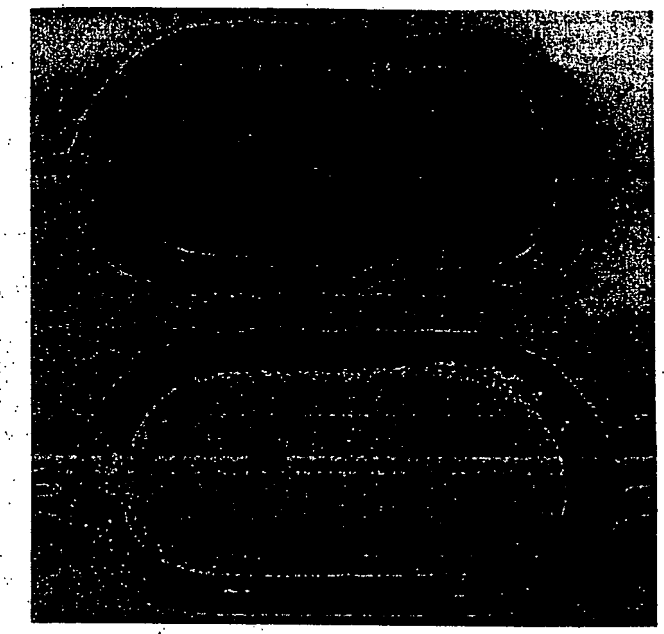


Figure 6.36. Manhole cover of a ball mill before (*top*) and after grinding (*below*).[2]

rial to be crushed. Adhesion is largely affected by molecular forces; however, partial melting and sintering are also possible.

Agglomerates are formed in the freely moving charge of a tube mill by the compaction of fine particles between the grinding media. Adhesion is affected by van der Waals forces between the particles that have been compressed very tightly. Beke,[4] who determined structural changes in the agglomerated particles, went so far as to regard this mechanism as similar to cold welding. Since these agglomerates are very strong, a so-called "grinding equilibrium" is obtained which has been observed and described by many authors.[5-9] It means that, after a certain grinding time, a state of equilibrium occurs, from that point on agglomerates are crushed during further grinding and reformed so that the apparent

Since every form of agglomeration decreases the efficiency of grinding and the degree of fineness obtained at the "grinding equilibrium" is not sufficient for many tasks, it is desirable to prevent or at least reduce these effects. In milling, one possibility to achieve less unwanted agglomeration is to add surface-active substances. It has long been known that small amounts of such additives may reduce the grinding time required for reaching a particular fineness by 20% to 30%.[10-14] Atoms or molecules of these substances that are present in a gas or vapor phase rapidly saturate free valences at the newly created surfaces which would otherwise give rise to recombination bonding. The effect of some of these grinding aids on the fineness of cement[15] after a specific grinding time is shown in Figure 6.37. It can be seen that, with

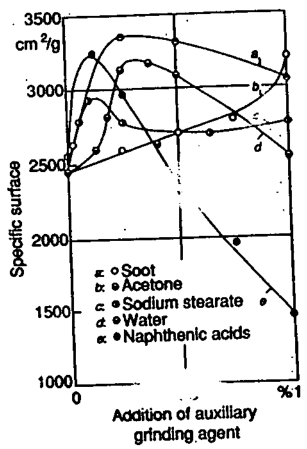


Figure 6.37. Effect of various grinding aids on the fineness of cement after a constant grinding time (2000 rev) in a rod mill (rod diameter: 25 mm, 85% critical speed). (According to Ghigi and Rabottino.[15])

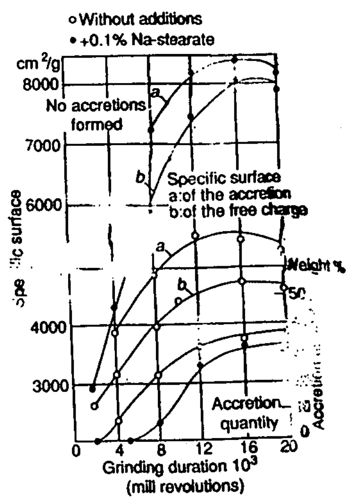


Figure 6.38. Specific surface of the build-up and of the free charge as well as amount of build-up with and without the use of a grinding aid (cement clinker, rod mill). (According to Ghigi and Rabotino.[15])

produced only if the amount of the grinding aid is very small. At higher concentrations the agglomeration tendency increases due to the formation of sorption layers and liquid bridges. In the case of soot a greater quantity is required because it is a solid wh[illegible] molecules are not very mobile. Good resu[illegible] also be obtained by merely enriching th[illegible]phere in the grinding chamber with ce[illegible]in gases or vapors that have been selected to possibly interact with the charge.[16–23]

As a rule, grinding aids also reduce caking. Figure 6.38 depicts the effect of 0.1% sodium stearate during the grinding of cement clinker. Other surface-active substances can delay build-up for longer periods or even prevent them entirely up to a certain fineness (for cement clinker, e.g., 0.1% triethanolamine,[4] Fig. 6.39). From Figure 6.38 it can also be seen that the specific surface, that is, the fineness of cement increases when 0.1% sodium stearate

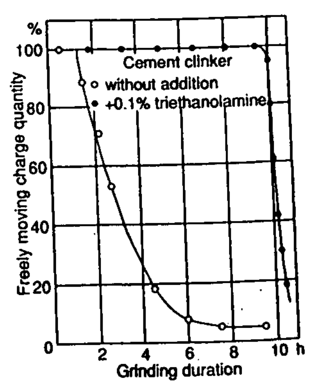


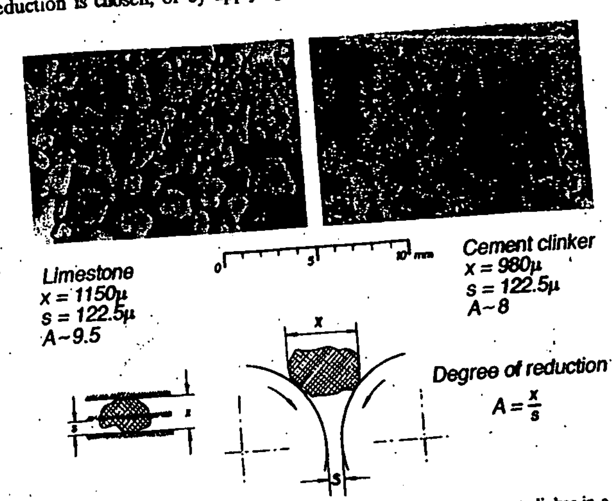
Figure 6.39. Changes in the amount of the freely moving charge during the grinding of cement in a laboratory ball mill with and without the addition of

The formation of lamellas or plate-like agglomerates in tube mills has been attributed to compaction occurring between the grinding media. The same mechanism happens in all comminution processes in which the material to be crushed is subjected to stresses by two surfaces. Since the second condition for the formation of agglomerates is a sufficient fineness of the particles, the occurrence of lamellas is observed mostly in fine grinding, for example, in roller mills.

One measure for the fineness as well as the intensity of stressing—and consequently, also for the agglomerative tendency—is the so-called degree of reduction, that is, the ratio of [illegible]imum feed particle size to the gap between the rollers. Figure 6.40 shows typical agglomerates produced in a roller mill with a high degree of reduction. Since the fine material is immediately compacted, almost all free valences at the newly created surfaces participate in recombination bonding.

Consequently, the formation of agglomerates can be avoided or reduced only if a smaller degree of reduction is chosen, or by applying friction between the rollers.[24] More recently it was found[25] that the combination of a large degree of reduction in high-pressure roller mills and the desagglomeration of the conglomerates produced by this method result in a significantly lower overall energy consumption during fine grinding of brittle materials (such as cement clinker and many ores); therefore, in many cases the unavoidable agglomeration of the fine particles is not only tolerable but the technology also results in a more economical fine grinding method.

Agglomerates can also be formed during impact grinding. Figure 6.41a shows schematically the fracture lines observed during impact stressing of a glass sphere.[26] A cone of fine material is created at the impact point and is compacted by the pressure resulting from the kinetic energy of the system into an agglomerated mass (Fig. 6.41b and c). Here too, the effect of free valence forces at newly created surfaces is utilized to its almost full extent, yielding a quite strong agglomerate. During impact crushing thermoplastic materials or organic substances with low melting points,



[illegible] [illegible]omerates produced during the grinding of limestone and cement clinker in a roller mill with a

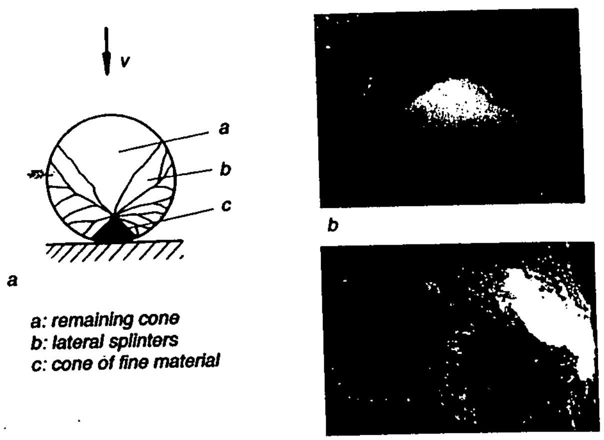


Figure 6.41. (*a*) Schematic representation of the fracture lines caused by impact stressing of a glass sphere.[26] (b) Agglomerated cone of fines created during the impact stressing of a glass sphere (impact velocity approx. 150 m/ sphere diameter 8 mm).[26] (*c*) Agglomerated cone of fines created during the impact stressing of a sugar cry (*left*).[27]

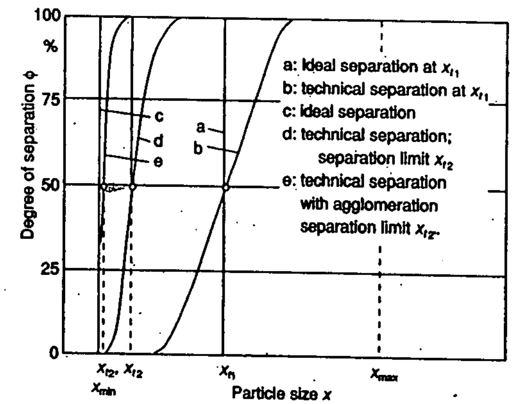


Figure 6.42. Qualitative representation of various separation curves.[2]

adhesion and agglomerate strength may further increase owing to melt bridges. It is very difficult to prevent such agglomeration; this can be affected only by reducing the impact velocity which, in turn, results in a lower degree of comminution. For glass spheres, for example, the formation of agglomerates was observed only at impact velocities exceeding 80 m/s.[26]

In wet grinding, as a rule, agglomeration is totally avoided by suspending the particles in liquid. Sometimes, the product of dry fine grinding is subjected to a brief final wet grinding to destroy the previously formed agglomerates.[50] Nevertheless, some materials also tend to flocculate in wet grinding. Since in these cases the adhesion forces are mostly electrical, the addition of a small amount of electrolyte nearly almost suffices to prevent flocculation.

Separation. During separation unwanted agglomeration can occur and needs to be avoided if a particle collective must be separated into

line. The so-called separation curve is a me sure of the quality of separation. In this cu the degree of separation (i.e., the percenta share of particles in the coarse and fine fr tion, respectively, after separation) is plot versus the particle size interval $x_{min} \leq x \leq$ which is to be separated. The cut size is t particle size of which half end up in the co fraction and half in the fine ($\phi = 50\%$). ure 6.42 is a qualitative representation of aration curves.[2] Line (*a*) in Figure 6.42 re sents the ideal or perfect separation at cu x_{t1} which is possible only in theory. In in trial separation equipment curves of the (*b*) are obtained. The sharpness of sepa increases with a steeper slope of the cu the abscissa uses a logarithmic gradatio aration curves representing similar sepa efficiencies at difference cut sizes are p to each other.

Agglomeration must be judged differ the separation task is to remove all p from a suspending fluid; then the cut

ly theoretically possible separation ality, a certain amount of smaller ains and the cut size is $x_{t2} > x_{min}$ If agglomeration occurs, the finest ay form larger entities or attach to les, changing the separation curve 42 to (*e*).

cases, however, agglomeration is ring the separation of particulate iques include screening, sifting, sorting, flotation, and, as a genl method, particle characterizacreening, agglomeration is often the motion of the material on pherical conglomerates are fred from material containing fines other adhesive characteristics. anisms are, for example: for solids, molecular forces and ads; for plastics, electrostatic ores, magnetic forces; for moist bridges and capillary forces; locking; and for materials with ts, partial melting and sinterstances several bonding mechr, simultaneously. In all cases paration by screening is dislomerated fines are classified les. Normally the effect of

agglomeration is reduced by mechanical destruction of agglomerates with, for example, rubber cubes placed on the screen decks, the application of brushes,[29,30] or the modification of amplitude and, respectively, frequency (ultrasonic screening)[31,32] of vibration. Agglomerates can be also destroyed by the effect of air jets passing through the screen from below.[33]

During the screening of moist bulk materials difficulties increase with moisture content but agglomeration tendencies are almost completely eliminated during wet screening when the particles are suspended in a liquid.[34] Since in moist screening particles or agglomerates are often retained in the mesh openings by liquid bridges, the separation of such materials is facilitated by direct electric resistance heating,[35] inductive heating,[36] or by altering the wetting angle and surface tension.[37]

In air classification, typically products from dry fine grinding are separated. Particular problems arise if the material to be separated contains agglomerates that were formed during comminution. Attempts are made to destroy these agglomerates by special designs of the feeder. Destructive forces are caused, for example, by sudden changes in speed or direc-

front of the classifier.[38] When classifying cement it was determined that grinding aids used during comminution also improve separation by avoiding agglomeration in the classifier.

In the classifier itself agglomerates are formed by molecular forces that may be reinforced by adsorption layers if separation is carried out in a moist atmosphere, by liquid bridges if moist materials are processed, and by electrostatic forces in a dry environment. Figure 6.43 depicts various separation curves of air classifiers.[39] With decreasing particle size the amount found in the coarse fraction increases, which is due to agglomeration whereby fine particles adhere to larger ones and conglomerates of fines behave like coarser particles. Both effects reduce the separation efficiency and can be avoided only if the causes of adhesion are removed, that is, mostly by eliminating moisture and humidity.

Sorting processes that separate materials according to particle characteristics other than size are mostly carried out in liquids. During a special technology, flotation, the relative capacity of material to float is enhanced by the addition of chemicals. Agglomeration can also reduce the separation efficiency of these processes because fine particles stick to larger ones, form conglomerates, or adhere to foam bubbles.[40] By use of modified chemicals, processing of very dilute suspensions, or multiple separation steps efficiently can be improved.

During particle size analysis, in addition to screening, sifting, and counting, sedimentation methods are often used that produce unequivocal results only if the individual particles elutriate without influencing each other. For that reason very dilute suspensions are used. Nevertheless, it is possible that agglomerates form or already present conglomerates do not disperse completely. Therefore, dispersion aids are often added that reduce particle affinity; a large number of such additives is available.[41-44] The molecules of dispersion aids attach to the particles, eliminating polarities and/or reducing interfacial tensions.

In connection with particle size analysis, the importance of correct sample preparation should be stressed. Because agglomerates always incorporate a relatively large number of the finest particles, the result of particle size analysis may be incorrect if preexisting agglomerates are not destroyed or conditions prevail during measurement that promote agglomeration.

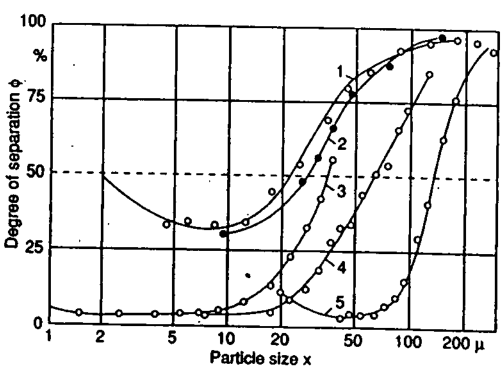
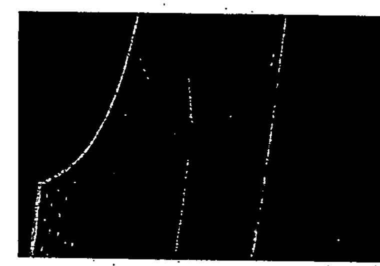


Figure 6.43 Separati...

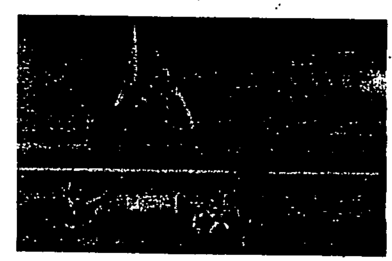
Mixing. Many of the previously mentioned considerations apply to the formation and prevention of agglomerates during mixing. Little needs to be added concerning mixing in liquids by stirring or methods for the production of suspensions and dispersions. The addition of dispersion agents is recommended when the tendency of the solids to agglomerate is high. Already present agglomerates or flocs can be destroyed by shear forces in the liquid. Consequently, the generation of the highest possible shear gradient is considered advantageous when selecting agitators.

During extended storage the particles in pharmaceutical suspensions often form agglomerates that can no longer be destroyed by shaking the preparation. This problem can be avoided by controlled flocculation of the solids.[45] After the addition of an electrolyte the fine particles aggregate to loose flocs that can by easily redispersed by shaking.

When mixing dry or moist bulk solids, agglomerates are formed, originating from the finest components of the mixture, which are held together by molecular and electrostatic forces as well as by capillary forces, particularly if the materials are moist.[46] These undesired agglomerates are broken up by shear or frictional stresses generated by the motion of the bulk mass or by special disintegration devices built into the blender. Figure 6.44 shows two examples. It depicts interior sections of a drum mixer with plow-like mixing elements and additional friction plates (a) or rapidly rotating cutter heads (b).



(a)



(b)

Figure 6.44. Partial views of the interior of a mixer with plow-like mixing elements. (*a*) With friction plates, (*b*) with cutter heads for desagglomeration.

Conveying. Particulate solids, especially finely dispersed powders, tend to form agglomerates and (sometimes thick) coatings on walls during conveying. Whereas agglomerates occur mostly on vibrating or shaking conveyors, wall build-up is more common in pneumatic conveyors.[47-49] The main causes of agglomeration during conveying of fine particulate solids are molecular and electrostatic forces. However, as a result of additional mechanical and thermal energy input, other binding mechanisms can also be activated, for example, partial melting and solidification.

Although it is very difficult to avoid the shaking conveyors several possibilities exist for the prevention of wall build-up and deposits during pneumatic transport.[47] For the latter, one of the most important conditions is to provide smooth inner wall surfaces to avoid the most common reason for initial build-up, the adhesion of the smallest particles in the roughness depressions. Since high drag forces will tend to remove particles that have already adhered to the walls, high transportation velocities will reduce the danger of build-up. For the same reason deposits will start in dead or calm areas of the system; therefore, such designs must be avoided. On the other hand, sudden changes in the direction of flow will cause high-energy impacts of particles with the wall, causing build-up. Finally, friction between particulate solids and pneumatic conveyor walls can ...

large extent on whether the particles and/or walls are electrically conductive or insulators. System design must take this into consideration.

Some results, published by Möller,[50] shall be reported to illustrate typical features of pneumatic conveying systems. The investigations were carried out during the transportation of particulate matter in a horizontal tube, 58.51 m long and 0.7 m in diameter. The pressures within the system could be determined at seven locations distributed along the measured length of the tube. $\Delta p = p_1 - p_7$ is the total pressure drop in the conveying system.

In Figure 6.45 the pressures at three different locations—1, 2, and 5—are plotted versus time. Since a fan located behind the dust collector at the end of the conveyor generates a slight negative pressure in the filter housing, a small negative pressure can be measured as long as the tube is clean. After a few seconds, however, the pressure p_1 rises and the other locations follow after short delays. Part of the pressure increase is caused by loading the air with particles, but a major portion is due to depositions building up in the tube. When the tube was inspected following runs of 20 and 50 s, respectively, no deposition was found in the first case, but after the longer run deposits had build up in the feed end portion of the system while the other parts still remained clean. Figure 6.46 depicts the total pressure drop between both ends of the tube. The lower diagram represents results of the same test as shown in Figure 6.45. After about 2.5 min the total pressure drop in the system remained almost constant. This indicates that, at least macroscopically, no further deposition takes place after this time.

The upper diagram in Figure 6.46 represents a completely different behavior. The total pressure drop increases more slowly. This is mostly due to the lower solid/fluid ratio, m_p/m_f (1.58 kg/kg as compared with 40 kg/kg) and the higher velocity (18.65 m/s versus 2.11 m/s). At rather regular time intervals, however, a high-pressure peak had been measured that was first observed at the feed end and propagated in a few seconds to the

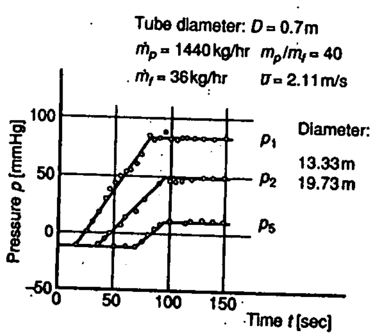


Figure 6.45. Pressure changes at three locations of an experimental pneumatic conveying system during the

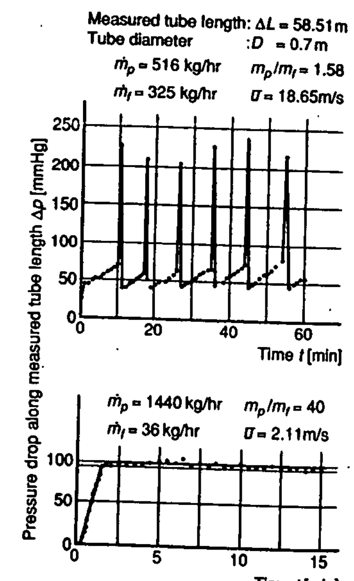


Figure 6.46. Pressure drop along the measured tube length of an experimental pneumatic

discharge end. This, together with some other observations, indicated that deposits fell off and were carried along, thus increasing momentarily the pressure drop. When the system was opened immediately after such a pressure wave went through, the inner walls were almost completely clean. The pressure drop curve shows further that the adhesion tendency is about constant for a given material and a conveying system operated at uniform conditions.

At high conveying velocities or in vertical tubes deposits build up uniformly. Such depositions shall be called "crusts" in the following. Whereas in the upper part of a horizontal tube, for instance, bonds between particles and walls are stressed by the weight of the deposit, they are strengthened in the lower part of the tube by gravitational forces. Therefore, especially in conveying at low velocities and high solid/fluid ratios in horizontal tubes, a second type of deposit is observed that shall be called "massy" deposit. Figure 6.47 describes schematically the formation of such deposits;[50] they are affected by gravity, grow in the direction of the mass flow, and are

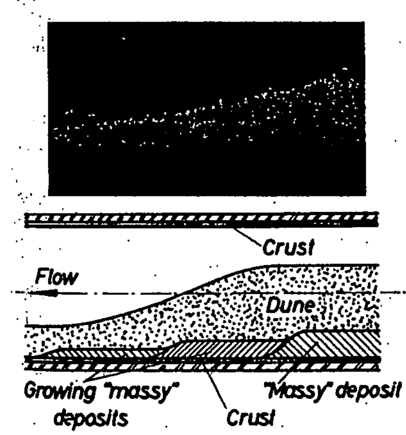


Figure 6.47. Sketch and photograph[50] of a model ex-

composed of finer particles because these particles exhibit higher adhesion tendency and because of a "sieving" or classification effect taking place in the charge by which finer components move to the bottom layer of the moving mass. The right part of Figure 6.47 is a photograph taken during a model experiment. With the exception of the formation of a crust, all other stages, including a "dune" of freely mobile particles moving over the deposits, can be distinguished clearly. Figure 6.48 is a view into a tube after pneumatically conveying a slightly moist quartz powder (particle size 50 μm) showing the heavy build-up in "crust" and "massy" deposit as well as the remainder of a "dune." Massy deposits can also be caused by the action of other forces such as centrifugal and inertial forces at an elbow.

Another important agglomeration phenomenon, which can be explained by the fact that the separation or dray forces define adhesion, is the controlled deposition yielding a more effective shape of the flow channel. Particles build up preferably in zones without flow or where the direction of flow lines is changed, such as by eddies, for example. A typical example of such deposits is shown in Figure 6.49. On the left, a partial cross-section of a "Möller pump" is presented; these pumps are used for feeding powders into a pneumatic conveying system. Powder and air enter a mixing chamber through a screw conveyor and a nozzle, respectively, and are then forced into the piping of a pneumatic conveying system. The photograph in the right part of Figure 6.49 shows a view (in direction A–A) of such a mixing chamber which was opened after conveying zinc oxide. Opposite the nozzle a deposit was built up forming a Venturi-like shape, which defines the most effective flow channel at this point. Similar depositions often can be found in pneumatic conveying systems that were not optimally designed and/or arranged.

Storage. Adhesion phenomena cause bridging of particulate solids in hoppers. In the case

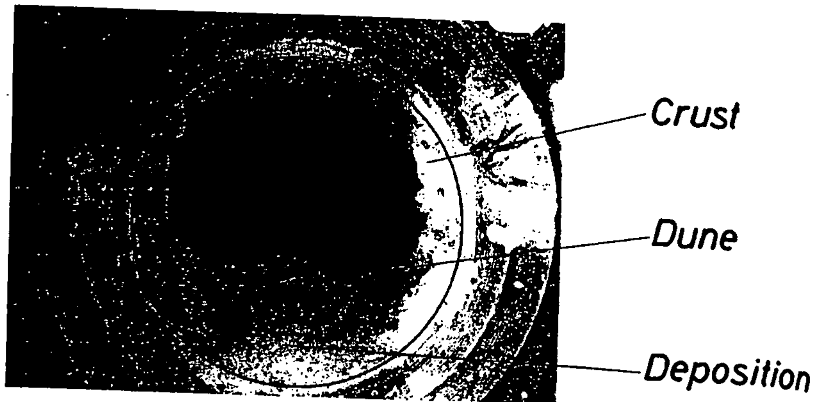


Figure 6.48. View into a tube of a horizontal pneumatic conveyor after conveying a slightly moist, finely divided quartz powder at low velocity.

is caused by building dome structures supported on the inclined walls in the lower, conical part of the bins.[51,52] With decreasing particle size, the participation of true adhesion forces in bridging and agglomeration increases. Bonding mechanisms are molecular forces and adsorption layers or liquid bridges. The latter often play an important role whereby liquid collects at the coordination points by capillary condensation.[54,55] Bridging can totally block the discharge from silos, thus causing severe operating problems. Because adhesion of finely dispersed solids cannot be avoided agglomerates and bridges must be destroyed by special devices. For this purpose, inflatable cushions are mounted in the hoppers or the material is momentarily fluidized by the injection of (pulsed) air jets. In the case of coarser solids, which tend to form domes, it is often sufficient to select a cone with steeper walls (="mass-flow" design). Small, remaining flow problems due to adhesion can then be overcome by installing vibrators or "hammers" on the outside silo walls.

Undesirable agglomeration is often observed if the particulate material is soluble or if chemical reactions can occur between the particles, particularly in the presence of moisture. These phenomena are very common in the fertilizer industry and are called caking if they occur in bulk masses or bag-set if the contents of the bags solidify.

Caking of fertilizers[49] and other soluble materials has long been and still is a great problem to producers and consumers of such materials. To get an idea about the importance and scale of this problem, three examples shall be presented at the beginning. Figure 6.50 shows the unloading of a shipload of sylvite that was expected to arrive as a free-flowing particulate mass but caked badly during transportation. Owing to the limited room in the shiphold the very costly and time-consuming method of manual unloading had to be chosen. Figure 6.51 shows the recovery of nongranular triple superphosphate from a curing pile which had to be blasted to break the so-called pile-set. This photograph, taken in 1947 by TVA, has historical value for this company because modern granular products, obtained by wanted, controlled agglomeration, no longer cake to such an extent that they require blasting. But, since especially high-nitrogen fertilizers are extremely hygroscopic, they must still be stored in bulk storage facilities with controlled, low humidity to prevent ...

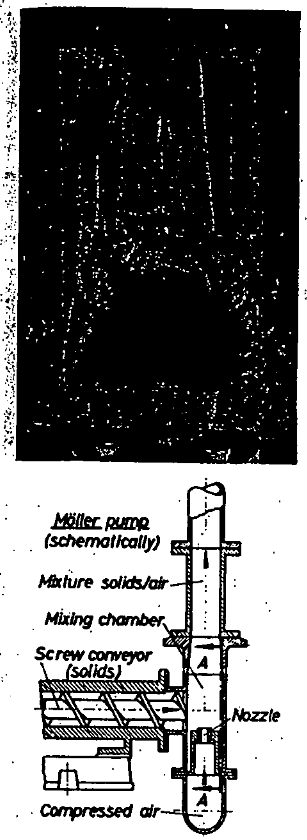


Figure 6.49. Sketch and photograph of a "Möller-pump." The photograph shows Venturi-like deposits after conveying of finely divided zinc oxide.[49]

The third photograph (Fig. 6.52) shows an example of the difficulties confronting the end-user. The granular fertilizer in the left bag displays the desired free-flowing behavior while the same fertilizer is caked in the other bag (bag-set). Even if such a caked mass is mechanically broken up, it will often no longer exhibit the same uniform conditions as an uncaked fertilizer and, hence, will negatively ...

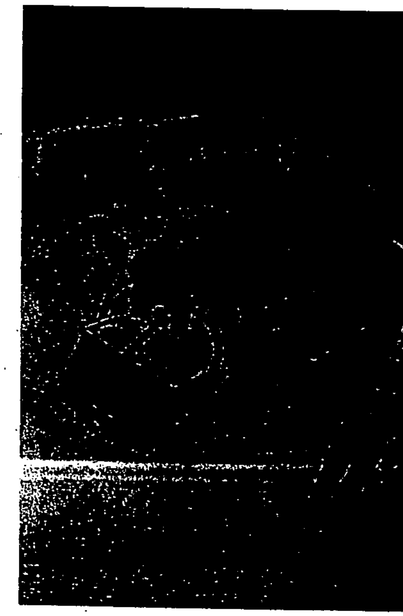


Figure 6.50. Manually unloading a shipload of caked sylvite.

Different materials become caked during various storage and handling procedures but caking itself is almost exclusively caused by solid bridges or, more specifically, by chemical reaction and crystallization of dissolved substances. Other binding mechanisms contribute only slightly to caking.

The rate and extent to which caking takes place depend on the moisture content, the particle size, the pressure under which the material is stored (e.g., top or bottom of the pile), the temperature, and its variation during storage, as well as the time of storage. The effect of these factors changes with different materials. Figure 6.53 depicts results obtained by Adams and Ross[56] in their "caking bomb." It can be seen that the crushing strength of caked masses rises with increasing moisture content (curves a in Fig. 6.53), caking

Figure 6.51. Recovery of nongranular triple superphosphate from a curing pile after blasting to break "pile-set."

temperature and temperature variations depends on the solubility. Figure 6.54 shows four typical temperature–solubility curves. Whereas the solubility of sodium chloride changes little with temperature, this is not true for potassium chloride and potassium nitrate, for example. The latter especially shows a very steep curve. Some salts, such as sodium sulfate, exhibit various temperature-dependent solubility ranges.

If salts or mixtures of different salts, such as fertilizers, for example, contain only a small amount of moisture, they can cake during storage or transport even in airtight bags if they are exposed to changing temperatures. In many cases (see Fig. 6.54) more salt will be dissolved if the temperature increases; this recrystallizes and forms solid bridges between the particles when the temperature drops

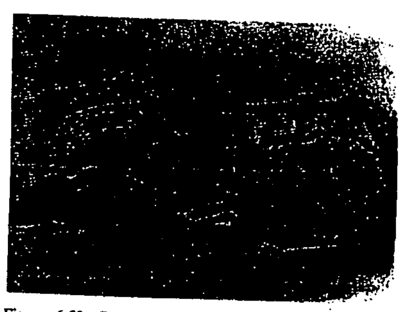


Figure 6.52. Granular fertilizer treated with an anticaking agent (*left*) and untreated control (*right*) showing severe "bag-set."

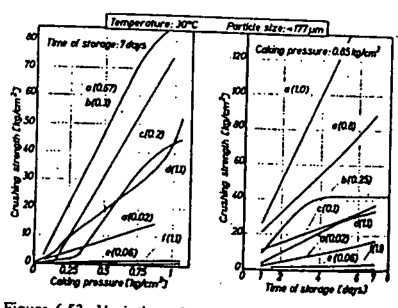


Figure 6.53. Variation of crushing strength with caking pressure (*left*) and time of storage (*right*). (*a*) $NaNO_3$, (*b*) $(NH_4)_2SO_4$, (*c*) urea, (*d*) KCl, (*e*) $(NH_4)H_2PO_4$, (*f*) superphosphate. The numbers in brackets indicate the respective moisture contents in percent.

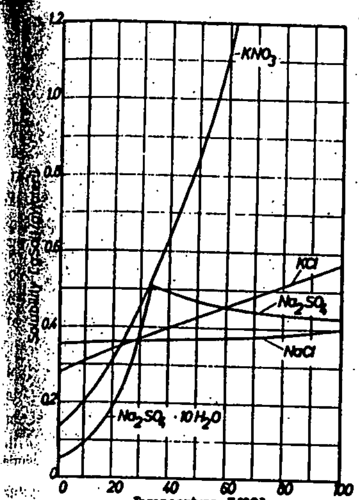


Figure 6.54. Solubility curves of four different salts.

again. Repeated cycling, for instance due to climatic changes or differences in day and night temperatures, tends to reinforce this bonding, causing bag-set.

The crushing strength of caked materials depends also on the number of bridges formed per unit volume and, therefore, decreases with increasing particle size.

In conclusion, it can be stated that the tendency for caking of a fertilizer mixture, for example, will vary with the physical and chemical properties of the components and their proportions in the mixture. It also depends on the method of mixing, the particle size after processing, and the storage conditions to which the finished products are exposed.[56–58]

The answer to what can be done to avoid or at least lessen caking is the same as in all other cases where unwanted adhesion or agglomeration must be prevented: Detect the binding mechanisms involved and the influencing parameters and then try to reduce their effect. In the following some examples shall be discussed.

(a): If (unobjectional) chemical reactions between components of a mixture do occur, these components should be mixed separately until the reaction has taken place. This intermediate product can then be blended with the other components and no longer induces caking. An example for this is any mixture that contains both ammonium sulfate and superphosphate.

(b): An almost trivial precaution is very often the lowering of the moisture content. However, this is not always necessary. Different maximum moisture levels exist that depend on the materials. Figure 6.53 shows that the crushing strength of superphosphate containing 1.1% moisture is very low while the strength of some other materials is much higher although they contain considerably less water.

Silverberg et al.[58] found during microscopic studies of several types of high-analysis fertilizers that caking usually resulted from bonding by the crystals of soluble salts. These crystals often covered the entire granule in the form of a veneer or hull. Figure 6.55 shows typical granular 12–12–12 fertilizer made with an ammonia–urea solution after 3 months of storage. They were illuminated from below and photographed at a higher magnification to reveal details of the crystalline hull. Bonding-phase salts identified during the study were potassium nitrate, ammonium chloride, monoammonium phosphate, ammonium nitrate, and an urea–ammonium chloride complex that are all highly soluble. Those salts migrated to the surface of the granule, leaving numerous small cavities within.

This mechanism needs water and drying should, therefore, reduce caking. Figure 6.56 is a photomicrograph taken with crossed Nicol prisms. It shows the difference in hull thickness between undried and predried 12–12–12 grade fertilizer granules. The crushing strength after storage decreased correspondingly.

(c): Curve *b* (ammonium sulfate) in the right-hand side diagram of Figure 6.53 shows the typical behavior of materials that respond

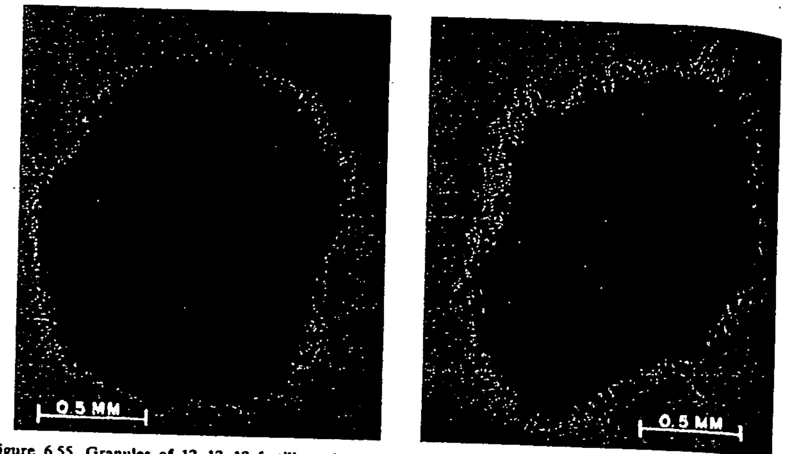


Figure 6.55. Granules of 12–12–12 fertilizer showing typical crystalline hulls of an urea–ammonium chloride complex after storage for 3 months in bags. (*left*) Uncured; (*right*) cured for 7 days prior to bagging.

prior to bagging. Such products cake in a few days to their final strength but the resulting lumps are broken up before the cured materials are finally bagged and put in storage. Curing can even accelerate hull formation as defined by Silverberg et al.[58] owing to the retention of heat and moisture in the pile. In products that respond well to curing, hull formation is apparently almost completed after curing and there is not sufficient additional development of crystals during subsequent storage to cause strong caking.

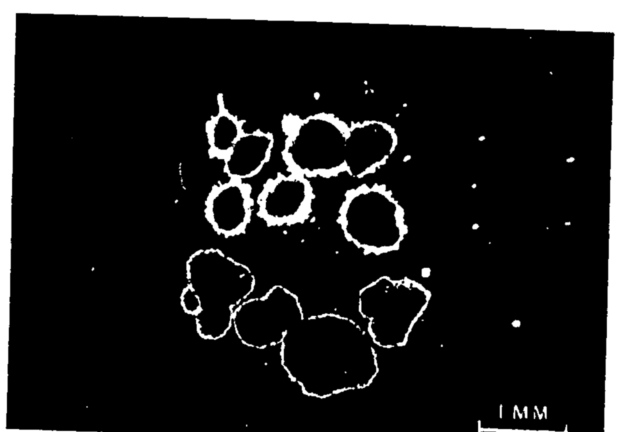


Figure 6.56. Difference in the hull thickness of undried (*top*) and predried (*bottom*) 12–12–12 granular fertilizer made with ammonia–urea solution.

Many products, however, do not improve during this type of curing. Figure 6.55 shows a comparison of uncured (left) and cured (right; 7 days prior to bagging) 12–12–12 fertilizer made from ammonia–urea solution, ammonium sulfate, superphosphate, potassium chloride, and sulfuric acid.[58] Although ammonium sulfate is present, the caking behavior of the other components dominates and both cured and uncured materials show continued growth of the hulls and caking during storage. Another curing method will be described under (e).

(d): The oldest method of "conditioning" fertilizers is the coating with a parting agent.[59,60] Storage properties are improved after addition of up to 3% of an extremely finely divided particulate solid, such as diatomaceous earth, kaoline, vermiculite, pulverized limestone, magnesium oxide, and a variety of other inexpensive powders. Siverberg and associates' microscopical studies[58] revealed again the fundamental properties of the "conditioner," which are threefold:

1. The powder coating the granule acts as a separator between the individual fertilizer particles and prevents intergrowth of crystals during and after granule formation.
2. The hulls form beneath the coating of conditioner and crystals seldomly project beyond the layer of conditioner.
3. The moisture is distributed uniformly over the surface of the granulae due to the high sorptive capacity of the finely porous coating. Thus, the localized growth of crystals at the coordination points is prevented and the surface hulls are much finer grained, more intergrown, and more densely packed than those covering unconditional products.

Such anticaking conditioning agents are usually applied by mixing them with the fertilizer in a rotary tumbler (typically a drum) prior to bagging.

(e): A modern variation of the above-mentioned conditioning process is the coating with surface-active organic chemic[illegible]

found, however, that not all surfactants improve the physical conditions of mixed fertilizers. Kumagi and Hardesty[62] reported that caking tendencies of mixed fertilizers were decreased by as much as 45% if nonionics were used but increased by as much as 37% with the use of anionics. Where in the process the surface-active agents were added was also found to be of decisive importance.

Typical cationic anticaking agents are fatty amines, for example, "Armoflos."[63–68] These amines, the general formula of which is $R{-}NH_2$ with R representing C_{16} and C_{18} chains, are believed to attach directly to the fertilizer particles with their surface-active amine group. Then, the fatty, hydrophobic part of the molecule extends outward from the surface, thus preventing hygroscopic products from attracting moisture. This is, of course, true only if a monomolecular layer covers the fertilizer particle and all amine molecules extend their hydrophobic portion outward. Therefore, too much conditioner will cause rather than prevent caking owing to the alternately hydrophobic and hydrophilic properties of additional layers.

This makes a modified curing process advantageous. The molecules of a second molecular layer, if attached, would position themselves with the amine group extending outward. These amine groups are fee to interact with other fertilizer particles, especially the phosphate portion of incompletely coated granules, to form an amine–phosphate salt. Pressure intensifies this effect. The chemical "bridge" is not as strong as the crystallized salt bridge and the "set" can be broken easily. Since, on the other hand, the amine–phosphate bond is stronger than the R–R bond, a more uniformly covered product results from a short bin cure (1 to 2 days) which is unlikely to set or cake again (see Fig. 6.52, left side).

Sometimes a combination of the two types of conditioner is used. An example for this approach is finely divided kaoline treated with

(f): A last method, granulating is today almost obligatory, particularly for mixed fertilizers. Size-enlarged, granular fertilizers offer fewer coordination points per unit volume where solid bridges can develop. If the strength of the bridges is low anyway, as in the case of superphosphate with 1.1% moisture or monoammonium phosphate with 0.06% moisture (see Fig. 6.53), granulating alone is sufficient to prevent severe caking.

Most of the above examples data back quite some time to a period when the fundamentals of unwanted agglomeration in different industries were investigated and means to avoid these phenomena were developed. While this part of size enlargement by agglomeration is often very important, because its effects may result in considerable losses of production and profit, mostof the literature deals with the methods and equipment to produce agglomerates with beneficial characteristics. Therefore, it is a most important achievement that recently a book, entitled *Cake Formation in Particulate Systems*,[69] on unwanted adhesion phenomena was published. Griffith, the author, distinguishes four major classes of particulate caking:

- Mechanical caking
- Plastic-flow caking
- Chemical caking
- Electrical caking.

In addition, several subclasses are defined whereby certain properties of components, either pure substances or part(s) of a formulation, can be expected to cause caking under certain conditions.

The chapters of the book describe the above, the chemistry of cake formation, phase behavior and cake formation, and electrically induced cake formation. Considerable emphasis is then given to laboratory techniques and test procedures that need to be considered by laboratories engaged in solving caking problems. Another chapter presents flow schemes to classify caked solids, an approach that is similar schematic or modern computer flow charts. On only 22 pages (out of 230 pages) the book then offers typical solutions to caking problems and concludes with a short chapter on induced cake formation, essentially a brief survey of what is called Desired Agglomeration in the context of this publication.

6.3.2.2 *Desired Agglomeration*

If size enlargement by agglomeration is carried out as a desired process the products resulting from this technology typically exhibit the advantages summarized in Table 6.5.[70]

Another somewhat different listing of benefits which, therefore, contained additional useful information, particularly "examples of application" was presented by C. E. Capes in Part 1 of Chapter 7 of the first edition of this book (Table 6.6).

6.3.3 Methods of Size Enlargement by Agglomeration

A common classification of methods for the size enlargement of particulate matter distinguishes between two types of processes:

- Growth/tumble agglomeration (no external forces)

Table 6.5. Advantages of Agglomerated Products.

1. No or low content of dust; therefore, increased safety during handling of, for example, toxic or explosive materials and, generally, fewer losses which may cause primary or secondary pollution
2. Freely flowing
3. Improved storage and handling characteristics
4. Improved metering and dosing capabilities
5. No segregation of co-agglomerated materials
6. Increased bulk density and lower bulk volume
7. Defined shape
8. Sometimes defined weight of each agglomerate
9. Within limits, porosity or density can be controlled; herewith dispersibility, solubility, reactivity, heat conductivity, etc. can be influenced
10. Improved product appeal

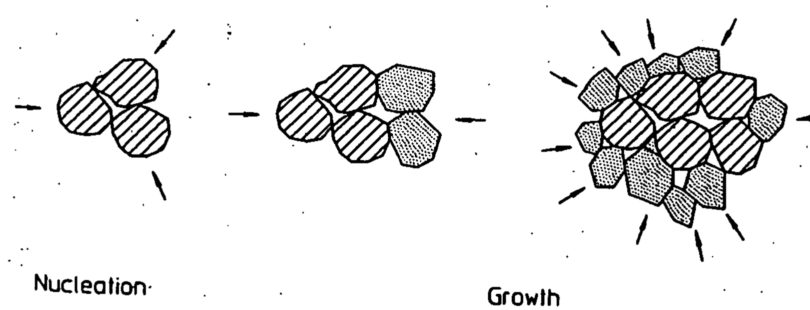
Table 6.6. Benefits of Size Enlargement and Some Representative Applications.

BENEFIT	EXAMPLES OF APPLICATION
1. Production of useful structural forms and shapes	Pressing of intricate shapes in powder metallurgy; manufacture of spheres by planetary rolling
2. Preparation of definite quantity units	Metering, dispensing, and administering of drugs in pharmaceutical tablets
3. Reduced dusting losses	Briquetting of waste fines
4. Creation of uniform, nonsegregating blends of fine materials	Sintering of fines in steel industry
5. Better product appearance	Manufacture of fuel briquettes
6. Prevention of caking and lump formation	Granulation of fertilizers
7. Improvement of flow properties	Granulation of ceramic clay for pressing operations
8. Greater bulk density to improve storage and shipping of particulates	Pelleting of carbon black
9. Reduction of handling hazards with irritating and obnoxious materials	Flaking of caustic
10. Control of solubility	Production of instant food products
11. Control of porosity and surface-to-volume ratio	Pelleting of catalyst supports
12. Increased heat transfer rates	Agglomeration of ores and glass batch for furnace feed
13. Removal of particles from liquids	Pellet flocculation of clays in water using polymeric bridging agents
14. Fractionation of particle mixtures in liquids	Selective oil agglomeration of coal particles from dirt in water

[redacted]

- [redacted]

The mechanism of growth/tumble agglomeration is similar to that of natural agglomeration (Fig. 6.57). Because the particles to be agglomerated are larger, the particle-to-particle adhesion must be enhanced by the addition of binders (mostly water and other liquids) and the collision probability must be increased by



providing a high particle concentration. Such conditions can be obtained in inclined discs, rotating drums, any kind of powder mixers, and fluidized beds (Fig. 6.58). In certain cases, simple tumbling motions such as on the slope of storage piles or on other inclined surfaces are sufficient for the formation of crude agglomerates.[71]

In most instances, growth/tumble agglomeration processes yield first so-called green agglomerates after growing nuclei into larger, nearly spherical aggregates by coalescence and/or layering (Fig. 6.57). These wet agglomerates are temporarily bonded by the effects of surface tension and capillary forces of the liquid binder. While, occasionally, components within the green agglomerate naturally produce permanent bonding, for example, owing to cementitious reactions, in most cases post treatments consisting of all or some of the following processes: drying and heating, cooling, screening, adjustment of product characteristics by crushing and conditioning as well as recirculating undersized material are necessary to obtain permanent and final strength (see right-hand side of Fig. 6.58). The sometimes very large percentage of recycle must be rewetted for agglomeration and needs to pass again through the entire process, which often renders this technology uneconomical.

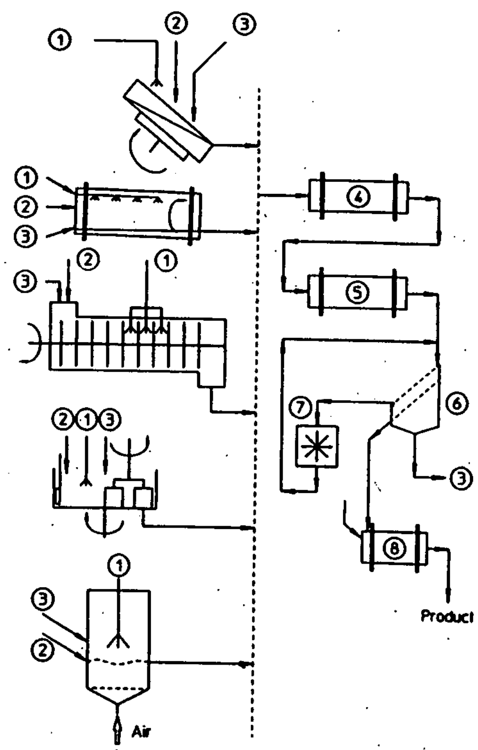


Figure 6.58. Schematic representation of typical equipment for size enlargement by growth tumble agglomeration.

With increasing size and mass of the particles to be agglomerated by growth/tumble methods, the forces trying to separate newly created bonds during agglomerate growth become larger until size enlargement by tumbling is no longer possible. Therefore, depending on the characteristics of the binder, there is a definite limitation to the coarseness of a particle size distribution which is in the range of x_0 between 200 and 300 μm. Considering the definition of x_0, the surface equivalent diameter representing the entire feed particle size distribution, it is, of course, possible to incorporate much larger particles, say up to approx. 1 mm, if a sufficient amount of finer grains is present in the mixture. Fine particles in the distribution will automatically influence x_0 through their large share in the specific surface area (see Table 6.4).

Relatively uniformly shaped and sized agglomerates can be obtained by [redacted] The moist, often sticky mass of [redacted] solids and a liquid binder is extruded through holes in differently shaped screens or perforated dies (Fig. 6.59). [redacted] Depending on the plasticity of the feed mix, short "crumbly," elongated "spaghetti-like," or cylindrical "green extrudates" are produced. In most cases a post treatment (typically drying and cooling) is required to obtain final, permanent strength. For the agglomeration of waste materials for recycling of beneficial use, only some of the medium-pressure "pelleting" equipment, particularly the design with flat die plate (Fig. 6.59, b.2), is applicable. As in the case of high-pressure ram extrusion (see below) this technique is particularly suitable for the agglomeration of elastic materials.

As far as applicability is concerned, high-pressure agglomeration (Fig. 6.60) is the most versatile technique for the size enlargement of particulate matter. If certain characteristics of

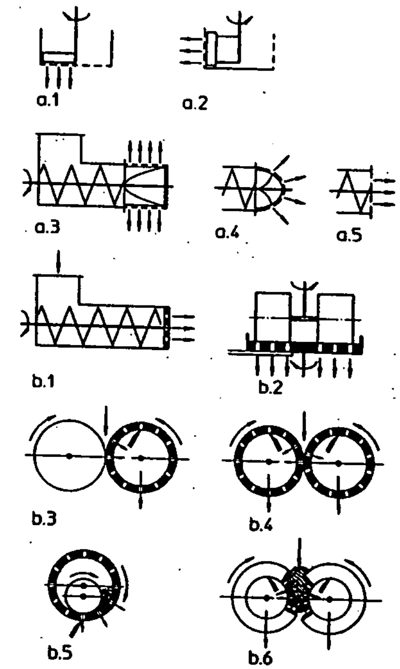


Figure 6.59. Schematic representation of equipment for low (*a*)- and medium (*b*)-pressure agglomeration.

during densification are considered during equipment selection, design, and operation, particulate materials of any kind and size (from nanometers to centimeters) can be successfully processed. [redacted] typically, the products from high-pressure agglomeration feature high strength immediately after discharge from the equipment. [redacted]

The mechanism of densification during pressure agglomeration [redacted]

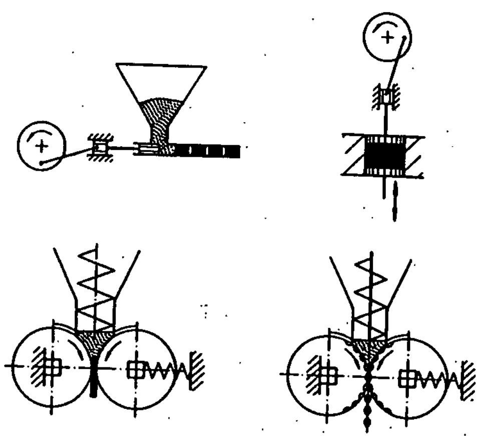


Figure 6.60. Schematic representation of equipment for high-pressure agglomeration.

little pressure followed by a steep pressure rise during which brittle particles break and malleable particles deform plastically (Fig. 6.61). Two important phenomena that limit the speed of compaction and, therefore, capacity of the equipment must be considered: compressed gas (air) in the pores and elastic spring back. Both cause cracking and weakening or destruction of the products from pressure agglomeration.

Compressed gas can be avoided if densification occurs slowly enough so that all air from the diminishing pore space is able to escape from the particulate mass and equipment. The problem becomes greater with finer particle size and requires special design features of the equipment. The effects of both phenomena, compressed air and elastic deformation, can also be reduced if the maximum pressure is held for some time (dwell time) prior to its release. Figure 6.62 shows that this is possible only with the ram extruder where all briquettes remaining in the extrusion channel are held at ~~a~~ certain pressure and redensified during each stroke. In punch and die presses a short dwell time can be achieved with some special drives whereas no such possibility exists with roller presses.

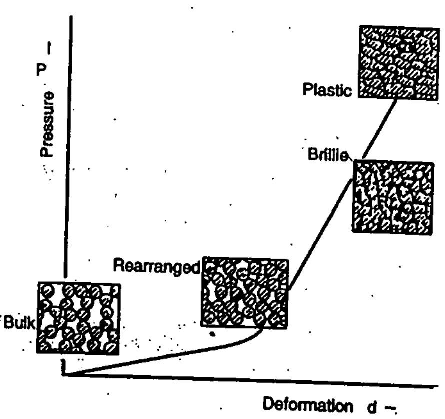


Figure 6.61. The mechanisms occurring during pressure agglomeration.

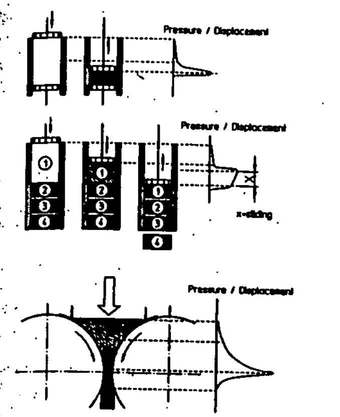


Figure 6.62. Pressure cycles in op~~en die (ram extruder), confined volume (punch and die press), and converging die (roller press) high pressure agglomeration.~~

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6.4 GROWTH/TUMBLE AGGLOMERATION METHODS—AGITATION METHODS

6.4.1 Introduction[1]

Growth/tumble agglomeration is the "most natural" of all size enlargement processes. As solid particles move in relation to each other in the relatively dense bed of a rotating or otherwise actuated containment of some kind or in a suspension with low solids density, particles will occasionally collide and, if the attraction or adhesion forces are high enough, coalesce. Theoretically, no specific piece of equipment is necessary for this phenomenon to occur; as long as the solid particles are kept in irregular, stochastic motion, the probability for collision and coalescence exists.

If in addition to this primary condition for agglomeration, the binding force remains strong enough to withstand the separating effects of all field forces and does not disappear with time without some other binding mechanism taking over, the "seed agglomerate" will survive and eventually collide with other single particles or agglomerates. At each instance of collision the above bonding criteria will be tested leading to either further growth, indifference, that is, the colliding partners will separate again and remain single, or conceivably destruction of weaker agglomerates.

For these adhesion criteria to test positive, the mass of adhering particles must be low and the specific surface high. This is equivalent to the requirement that the size of agglomerating particles must be small, typically in a range below approx. 100 to 200 μm. Micron and submicron particles (approx. <5 to 10 μm) will adhere to form an agglomerate even if they are dry. van der Waals forces are high enough to cause coalescence. Agglomeration of larger particles necessitates the addition of binders.

Drawbacks of all tumble agglomeration methods are the limitation to small dimensions of the particles forming the agglomerate and that, in most cases, only temporarily bonded conglomerates are formed. A curing step must follow to obtain permanent bonding which often also results in considerable strengthening of the agglomerate. In the green stage, the main binding mechanisms are bridges of freely movable liquids and capillary pressure at the surface of particle aggregates filled with liquid as well as adhesion caused by viscous binders; in the case of very small particles, field forces such as van der Waals, electrostatic, or magnetic attraction may also participate. After curing, agglomerate strength is achieved by solid bridges resulting from sintering, chemical reaction, partial melting and solidification, or crystallization of dissolved substances.

Tumble agglomeration equipment can handle large volumes of material effectively if the above criteria are fulfilled. The apparatus is simple and the design is unsophisticated. The expensive part of tumble agglomeration is normally the curing step of the process, which also contributes high operating costs. However, if very large amounts of solids must be agglomerated and the fine particulate form is also required for other reasons, for example, the concentration of valuable constituents of ores, tumble agglomeration is a preferred technology. In those cases the main binder is normally water. At capacities exceeding one million tons per year the curing facilities also become more economical and methods for, for example, heat recuperation to reduce operating costs are feasible.

Other reasons for the application of tumble agglomeration, even at small capacities, may be the high porosity of the agglomerates with other attendant beneficial characteristics such as large surface area (e.g., for catalyst carriers) and easy solubility [e.g., for food (drink) and pharmaceutical products]. These advantages may be so valuable that additional grinding costs to obtain the necessary small particle size for agglomeration will be accepted and high operating costs can be absorbed. In these cases even the agglomeration liquids (binders for the formation of green agglomerates) are sometimes so valuable that they are condensed from the dryer off-gas and recirculated.

6.4.2 Definitions[1]

With the exception of very few applications where particles are so small that they naturally agglomerate in the dry state, tumble agglomeration methods utilize binders. Even in those materials that contain the binder component inherently, this constituent of the bulk mass to be agglomerated is so obvious that one cannot classify such ...

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6.5 PRESSURE AGGLOMERATION METHODS

6.5.1 Introduction

Pressure or press agglomeration using tabletting machines and other piston presses, roller presses, isostatic pressing equipment, and [redacted], as well as some lesser known equipment, represents a large share among commercial applications of [redacted] This technology is largely independent of feed particle size and the forces acting upon the particulate feed may be very high with certain equipment. Therefore, it constitutes the most versatile group of size enlargement processes by agglomeration. Because of the relative complexity of the equipment and its comparatively small capacity per unit, these techniques find their largest field of use in low to medium capacity applications (approx. 1 to 50 t/h). In addition, specialty products, such as those in the pharmaceutical industry, may be processed in very small and sophisticated machinery, handling only a few kilograms per hour, while

ple, some fertilizers and refractory materials, are briquetted or compacted in large facilities employing multiple units.

Other advantages of pressure agglomeration are that [redacted] that the amount of material in the system is relatively small. Therefore, pressure agglomeration methods lend themselves particularly well to batch or shift operations and to applications in which several products must be manufactured from different feed mixtures. At the end of a production run, the system can be easily and completely emptied in a relatively short period of time.

In general, if several million tons per year of always the same feed composition must be agglomerated, such as in ore or minerals mining and concentrating, pressure agglomeration will normally not be the preferred first choice. In all other cases, one of the different methods of pressure agglomeration should be considered.

6.5.2 Mechanisms of Compaction[2]

The production of a powder tablet, compact or briquet can be carried out by a number of techniques, the purpose of which is usually to form the powder into a more or less well-defined shape. [redacted] However, all have in common a basic compaction mechanism.

When a particulate solid is placed into a die and pressure is applied, a reduction in volume will occur due to the following mechanisms (Fig. 6.110):

1. At low pressure, rearrangement of the particles takes place, leading to a closer packing. At this stage, energy is dissipated mainly in overcoming particle friction, and the magnitude of the effect depends on the coefficient of interparticle friction. In the case of fine powders, cohesive arches may

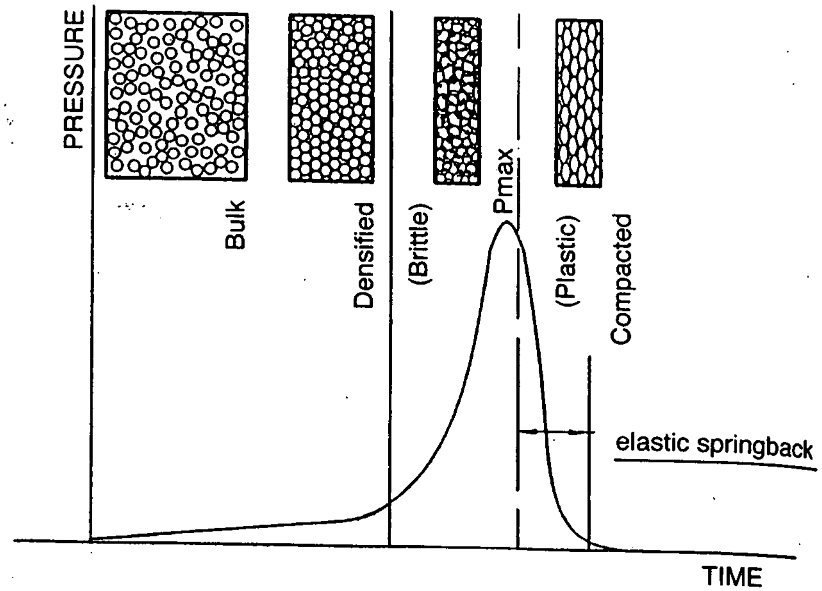


Figure 6.110. Mechanisms of compaction.

2. At higher pressures, elastic and plastic deformation of the particles may occur, causing particles to flow into void spaces and increasing the area of interparticle contact. Interlocking of particles may also occur. For materials of low thermal conductivity and low melting point, the heat generated at points of contact may be sufficient to raise the local temperatures to a point where increased plasticity and even melting facilitate particle deformation.

   With brittle materials, the stress applied at interparticle contacts may cause particle fracture followed by rearrangement of the fragment to give a reduced volume.
3. High pressure continues until the compact density approaches the true density of the material. Elastic compression of the particles and entrapped air will be present at all stages of the compaction process.

The mechanisms discussed may occur simultaneously. The relative importance of the various mechanisms and the order in which they occur depend on the properties of the particles and on the speed of pressing.

The aim of compaction is to bring small particles into sufficiently close contact so that the forces acting between them are large enough to produce a product that has sufficient strength to withstand subsequent handling. Therefore, it is often necessary to carry the compaction into the bulk compression stage, in which the stressing is hydrostatic in character. Broken or deformed particles are no longer able to change position because of the few remaining cavities, and a certain amount of interparticle conformity has been achieved. With increasing pressure the apparent density will gradually approach that of the theoretical density. The rate of this approach depends on the yield point of the material. Brittle materials are more difficult to densify to a high degree by pressure only because fragmentation decreases due to the hydro- ...

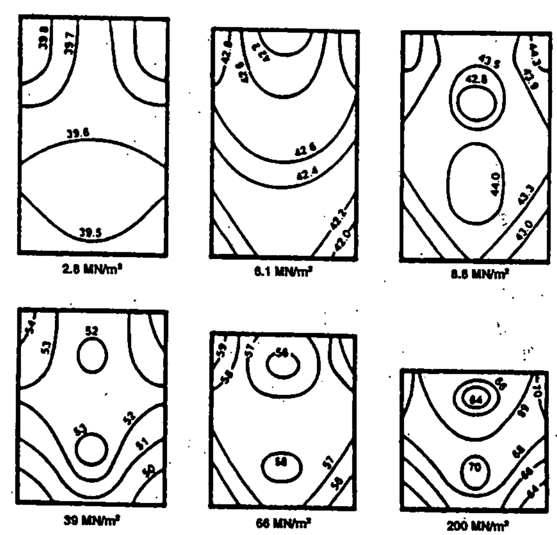
... smaller particles. When porosity becomes fully disconnected, the isolated pores may set up considerable internal gas pressures which, together with stored elastic energy, can contribute to the disintegration of compacts if the pressure is released too quickly.

If a particulate solid were compacted in a cylindrical die with frictionless walls, it is expected that the pressure exerted by the piston would be transmitted throughout the material giving uniform pressure and, therefore, uniform density throughout the compact. In practice, the presence of frictional shear forces at the wall leads to a nonuniform pressure distribution causing variations in the density of the compact (Fig. 6.111). These variations are present in products from all pressure agglomeration techniques and lead to weakening of the compact. If a sintering step follows, distortion is possible owing to differences in the amount of contraction occurring at the positions of different density.

Figure 6.111 shows density distribution curves in tablets produced in a cylindrical die with stationary bottom after one-directional compression (punch moves from the top into the die).[3] The individual tablets were obtained from identical bulk volumes after applying the indicated compaction forces. In such tablets the highest density is at the top edge of the compact and the lowest density at the bottom edge. A region of high density occurs near the axis a short distance above the bottom of the compact. In some cases the density in this position is higher than that observed near the axis at the top of the compact.

The general conclusion from investigations into the effects of operating conditions of pressure agglomeration equipment are that density variation:

- increases with the applied pressure and with the height of the specimen for constant diameter,
- decreases with increasing diameter even at constant height-to-diameter ratio,
- is slightly reduced by the addition of a lubricant to the powder, and



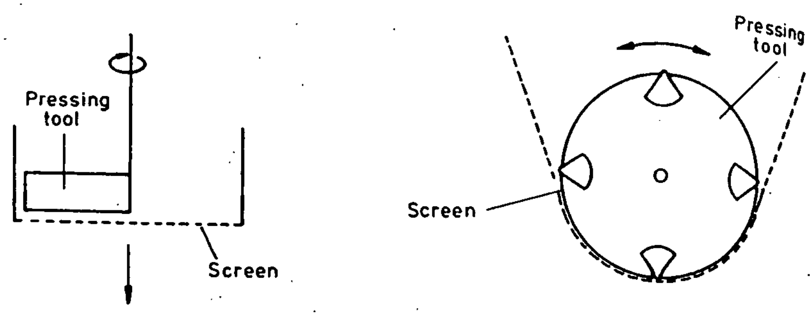


Figure 6.112. Schematic representation of two typical low-pressure agglomerators.[1]

- is considerably reduced by lubricating the die walls or tools.

Segregation during feeding and filling also leads to density variations owing to local changes in size distribution and, in the case of mixtures, to differences in the plasticity and friability of the component materials. Since there is evidence that radial flow of powder during compaction is negligible, it is expected that variations in density before compaction have an appreciable effect on the uniformity and quality of the compact.

A knowledge of the relationship between compacting pressure and density is important because pressure or force, more than any other factor, controls the attainment of high density, high strength, and low porosity in green compacts and markedly influences the same properties in the final product. A number of empirical formulas has been proposed to describe the pressure–porosity relationship; however, none of these formulas is universally applicable, giving acceptable results over a limited range of pressures only.

6.5.3 Low- and Medium-Pressure Agglomerators

6.5.3.1 General

Low-pressure agglomeration is most probably the oldest granulation method for particulate matter. Originally, a moist mass was passed through a sieve by the eminence of the hand, a spatula, specially designed handtools, or a brush. Later, this procedure was simulated by mechanization. Figure 6.112 depicts schematically two typical low-pressure (screen) agglomerators. The size of the screen openings depends on the moisture content of the mass to be agglomerated.

In most cases, the material pressed through the screen must be scraped off with suitable tools (knife blades). The green product is collected and dried. A typical system with continuous drying is shown in Figure 6.113. If necessary, all or only larger granules may be crushed to the desired size in a mill. The shape of the final, dry agglomerates produced by low-pressure agglomeration is slightly elongated

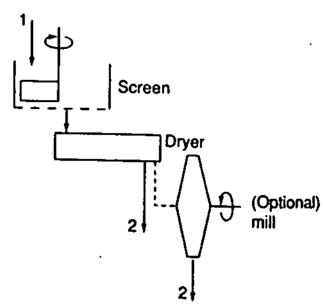


Figure 6.113. Low-pressure agglomeration system with screen, dryer, and (optional) mill. 1: Powder + binder, 2: granular product and fines.[1]

generally irregular and density is low (high porosity and solubility).

As far as porosity, solubility, and the possibility to introduce microdoses of active ingredients with the agglomeration liquid are concerned, for example, in the pharmaceutical industry, products from low-pressure agglomeration are similar to those obtained in tumble agglomeration. The main differences are that the particle shape is more irregular, particularly if all or part of the dried material is milled to adjust particle size, and that the steps of mixing, agglomeration, as well as drying are carried out in separate process equipment. The latter may be an advantage (better control of each step) or a disadvantage (possibilities of material losses, contamination, etc.) or both.

[redacted] peripheral axial, or dome discharge, low-pressure screen agglomerator (see Fig. 6.59, a3 to a5) or, for a denser extrudate, employ a medium pressure axial die plate (see Fig. 6.59, b1). [redacted] In case of low-pressure extrusion tapered rotors with longitudinal blades expel the material through a screen (Fig. 6.114), which is easily replaced or changed for different extrudate diameters. Screen openings as small as 0.5 mm are possible for many materials. For medium-pressure applications the peripheral discharge attachment is replaced with axial die plates.

Medium-pressure agglomerators use extrusion for the formation of agglomerates. In this respect the mechanism is similar to screen agglomeration in low-pressure agglomeration. To achieve higher densification, forces are created in thicker dies by friction of the material sliding through mostly cylindrical extrusion channels or bores. In agglomeration, this technology is called [redacted].

Schematic representations of the machines are shown in Figure 6.59, b1 to b6. The most commonly utilized equipment features differently arranged press rollers and perforated dies (see Fig. 6.59, b2 to b6). If the extrusion bores are long and without relief counterbores, relatively high densification can be achieved. On exiting, the [redacted] agglomeration techniques use moist mixtures, that are prepared in a mixing step prior to pelleting.

An important advantage of medium-pressure agglomeration is that, in comparison with tumble or low-pressure granulation, only

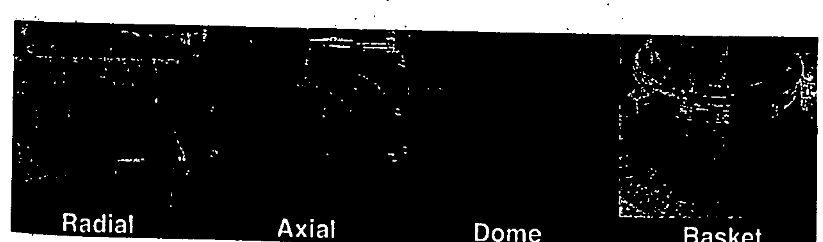


Figure 6.114. Photographs of low-pressure agglomerates exiting from screw extruders with radial, axial, and domed discharge screens. The basket-type (extruder) granulator is also shown.[4]

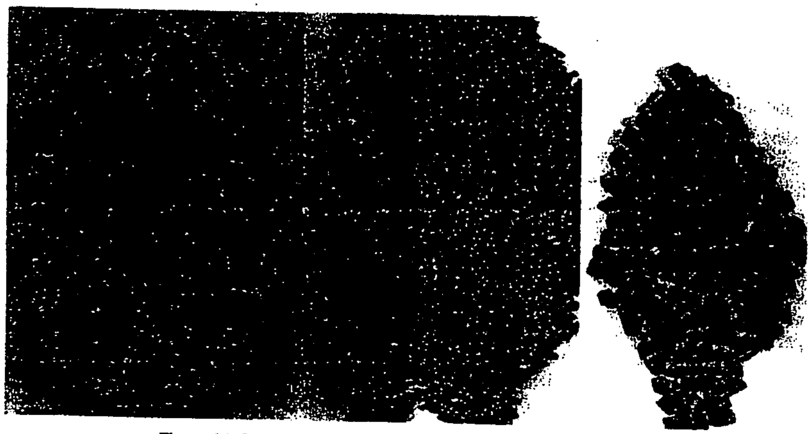


Figure 6.115. Typical products manufactured with a pelleting machine.

one half to one third of the agglomeration liquid is required. Therefore, drying takes place quicker and with less energy.

For mechanical reasons it is not easily possible to equip the dies with bores of less than 1 mm diameter. This is why agglomerates formed by medium pressure (extrusion) are normally dried and then "crumbled" by crushing if a finer granular product is desired. Fines may be screened out and recycled to the mixer for renewed agglomeration.

6.5.3.2 Equipment

Continuous Extrusion. The phenomenon of movement caused by the flights of rotating screws in more or less tightly fitting housings can be used to continuously produce the necessary pressure to overcome the friction in open-ended dies. These so-called screw extruders offer advantages compared with, for example, the noncontinuous ram extruders (see below) because capacity limitations due to the reciprocating movement of the plunger with its acceleration and deceleration phases do not exist. Feed and product move continuously, thus avoiding static friction, and additional work, such as plasticizing or even melting and deaeration or degassing can be performed by specially designed screws.

Screw extruders may feature single or twin screws. While most of the modern machines are used in the plastics industry to produce granular master compounds with complex equipment design,[5-8] relatively simple presses are utilized for agglomeration by extrusion of plastic and pasty materials such as clays, lightweight aggregate mixtures, building material mixtures, coal or carbon products with binders, etc.,[9] and of powders mixed with liquid binders and, sometimes, lubricants or plasticizers.

In general, the extrusion rate dm/dt of a screw extruder is determined by the combined influence of screw transport and die resistance. The operating point, defining pressure and capacity, is obtained in a mass flow/pressure diagram at the point of intersection between the lines characterizing the screw and die performance, respectively (Fig. 6.116). Because of the influence of both characteristics, the theory of screw extruders is rather complex. The actual operating condition results from the superposition of two extremes, of

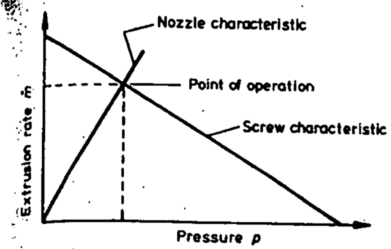


Figure 6.116. Extrusion rate $\dot{m} = dm/dt$ of a screw extruder as a function of pressure of the mass to be extruded.[5]

screw conveying with no backpressure and pumping/mixing against a completely closed end.

The above-mentioned difficulty in describing mathematically the conditions in a screw extruder becomes even more complicated if special kneading, densification, and deaeration sections are included in the design. As shown in Figure 6.117 the simplest single screw extruder features already three distinct zones: feed, transport, and compression/extrusion zones.

In some units, a conditioning mechanism is located in the feed zone so that liquid can be introduced followed by kneading of the wetted powder mass into a moist, homogeneous mass. Some mixing of different powders may also be accomplished.

The auger-like screws then transport the material into the compression zone, where air or gases are forced from the interstitial voids as particle matter is compacted.

Screw designs vary in accordance with how much pressure is needed to obtain sufficient densification and to overcome the friction in the die. In the space between the end of the screw and the die, densification is controlled by rheological properties of the material. Less compression and less dense extrudates are obtained if this gap becomes smaller and vice versa.

Extruders that rely solely on the pressure developed by the rotating screws employ hy-

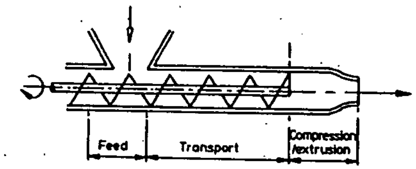


Figure 6.117. Operating conditions in a simple, axial, single-screw extruder.

drostatic pressure as the transport mechanism and are generally high-pressure extruders. Those extruders that utilize dragging or rolling motion feature a localized "drag flow" transport mechanism and, consequently, the rate of work performed and internal pressure developed are lower.

Two fundamentally different mechanisms for screw extrusion are possible: axial (Figs. 6.117 and 6.118a) and radial (Fig. 6.118b). Both machines may be equipped with either one or two screws.

While most of the axial screw extruders operate solely according to the hydrostatic pressure principle (Fig. 6.117) several other types use an extrusion blade to additionally create a wiping effect at the die plate (Fig. 6.118a). This blade looks like and performs in a fashion somewhat similar to a propeller. Nevertheless, the material discharges axially from the bores at the end of the extruder barrel.

In radial discharge extruders the extrusion blades are formed as shown in Figure 6.119. Material is extruded circumferentially through openings in the barrel wall and the direction of extrudate flow is perpendicular to the screw axis. In many cases, the barrel wall in the extrusion zone consists of a screen. Because of the extremely short length of the extrusion openings in such equipment, low-energy input and low densification prevail. Extrudates formed by this mechanism are very plastic and are normally treated in a second step, for example, achieve final shape and density.

As with all pressure agglomeration techniques, air or gases are squeezed from the

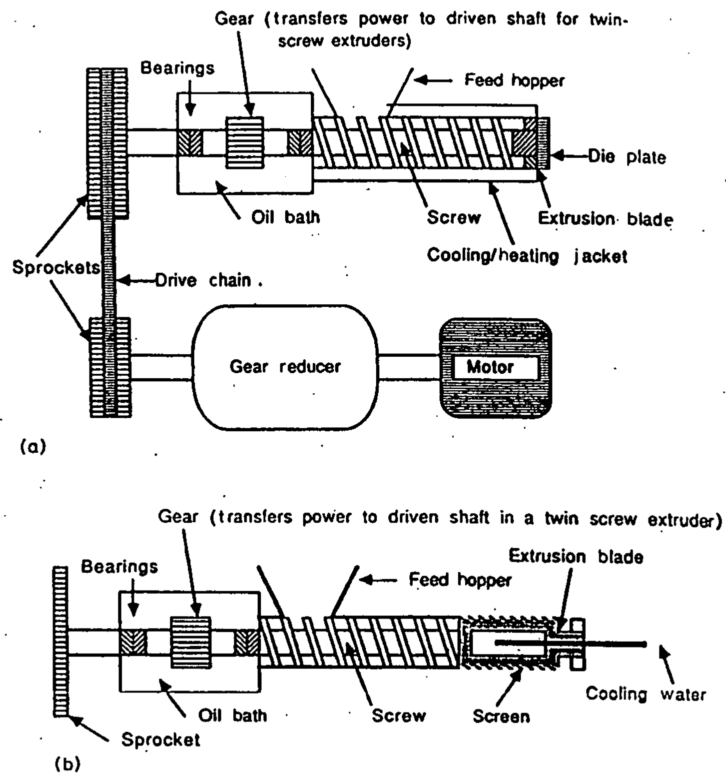


Figure 6.118. Schematic representation of (*a*) axial and (*b*) radial screw extruders.[10]

particle interstices during densification. The complete and reliable removal of this air or gas from the equipment is most important for good product quality. Because forward flow into the denser compression area and through the die opening is very restricted, air must normally flow in opposite direction of the flow of material and escape at the feed opening.

The product shape is defined by the shape and length of the opening in the die or screen. If a denser product is desired, a thicker die plate or screen is required to increase backpressure. If feasible in regard to product size, a similar effect can be obtained by reducing the diameter of the die opening. The lower unit of this dimension is defined by the decreasing economics of manufacturing the holes and the increasing backpressure due to a reduction of the free area (relatively higher amount of land area between the holes is required for structural reasons). The upper limit on hole size is determined by the flow properties of the particular formulation, the extrusion rate, and the ability of the extruder screws to transport and compress sufficient material so that a consistent extrudate is obtained. At the same time, relatively thick die plates are necessary.

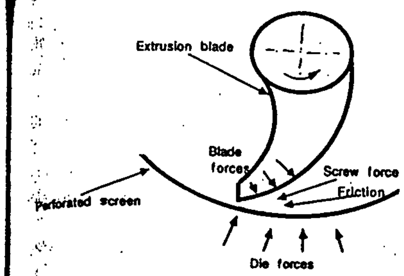
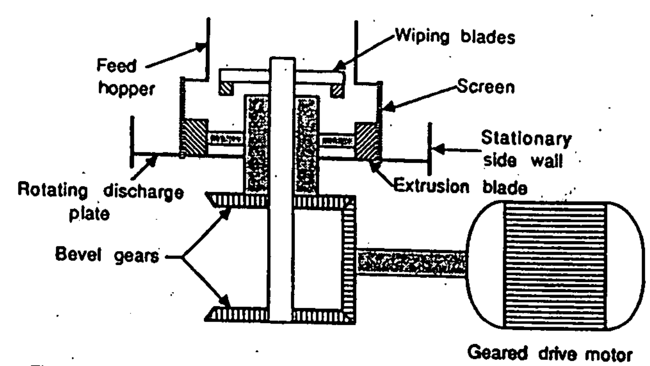


Figure 6.119. Extrusion blade and forces in radial screw extrusion.[10]

The difference between a screen and a die plate extruder is quite substantial. While die plates are 2 to 30 mm thick, screens feature usually the same thickness as the hole diameter; screens are rarely thicker than 1 mm. For extruders with the same barrel diameter, a radial discharge with screen will have more than 6 times the open extrusion area than an axial discharge with die plate.[10] This has consequences for screw design but will also generally translate into higher specific capacity and cost advantages for the radial discharge extruder if the low density and small product diameter can be tolerated.

Another continuous extrusion press that finds increasing but specialized application, mostly in the pharmaceutical industry, is the basket type extruder (Fig. 6.120).[10] This type of equipment is similar to the radial discharge extruder except that material is fed into the extrusion zone by gravity rather than screws. The perforated cylinder sits upright so that feed material falls into the basket and in front of rotating or oscillating extrusion blades with vertical axis of the rotor. The material is compressed in the nip between blade and screen and forced through the holes in the screen; the extrudates are transported to a discharge chute by a slowly rotating horizontal table. Forces developed in basket type machines are similar to those described for screw extruders except that the additional compressive force of the screw(s) is not present. These devices generally result in the least compaction of all extrusion apparatus and, therefore, the number of applications is rather limited. Attractive features for the pharmaceutical industry are: low energy input coupled with minimal temperature rise in the mass, high porosity, and quick dissolution of the product.

Power consumption, equipment geometry, wear rate, as well as capital and operating costs are all directly correlated to the internal working pressure. Therefore, there are good reasons to consider lower extrusion pressures obtained in peripheral or radial extruders and pelleting machines.



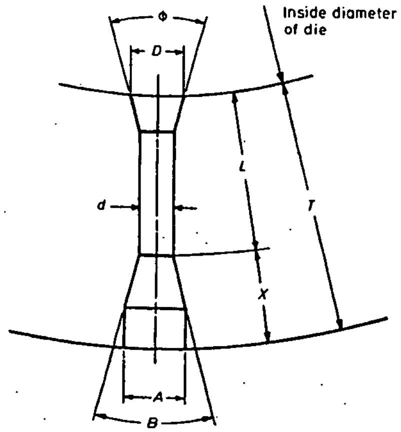


Figure 6.125. Die hole characteristics.[12]

and increasing the bore diameter d. Die life is determined by the increase of d to such dimensions that either the product size is no longer acceptable or the backpressure becomes too low (reduced compression and, therefore, inadequate product density and/or strength). If neither characteristic is critical, the limiting die life is defined by the remaining structural integrity of the die.

Particularly if pellets with high density or strength and small diameter must be produced, the necessary die thickness and effective hole length may be rather incompatible. In such cases replaceable insert plates with short bores may be used (Fig. 6.126a). Other inserts may be used as replacements in case of wear and to salvage the overall die body (Fig. 126b)[13] or to fulfill process requirements, such as cooling of the dies.[14]

Figure 6.127 shows two typical designs of pellet mills. Figure 6.127a is a partial cut through a machine with concave die rings and Figure 6.127b is a photograph of the working parts of a flat die pellet mill. For structural and process reasons the perforated concave or flat rings cannot be very wide (Fig. 6.128); therefore, to increase the capacity of a given press and more uniformly distribute the load, up to three rollers are installed in concave die presses (Fig. 6.129) and up to five rollers are used in flat die presses (Figs. 6.127b and 6.128b). Adjustable plows direct the feed in front of each press roller (Figs. 6.127b and 6.129), thus approximately increasing the capacity by the number of rollers used. Because in flat die pellet presses additional, potentially unwanted shear develops between cylindrical rollers and the die plate, relatively narrow rollers and perforated die areas are used. Press

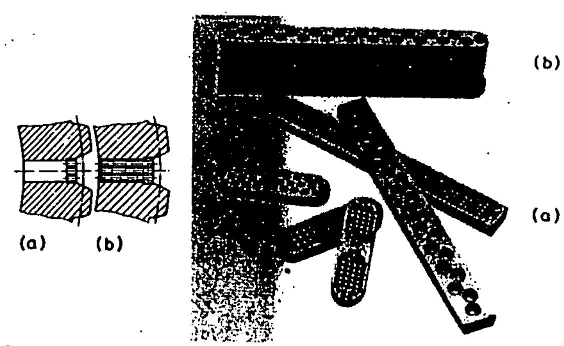


Figure 6.126. Replacement insert plates with (a) short and (b) long bores (BEPEX/Hutt, Leingarten, Germany).

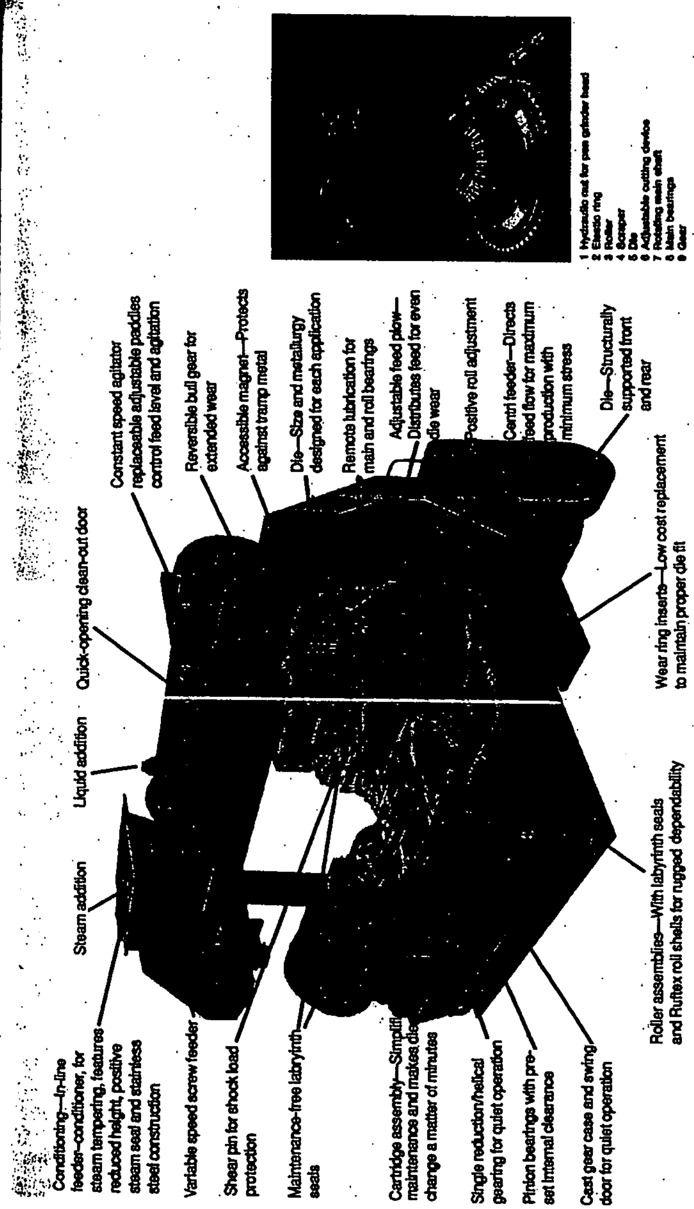
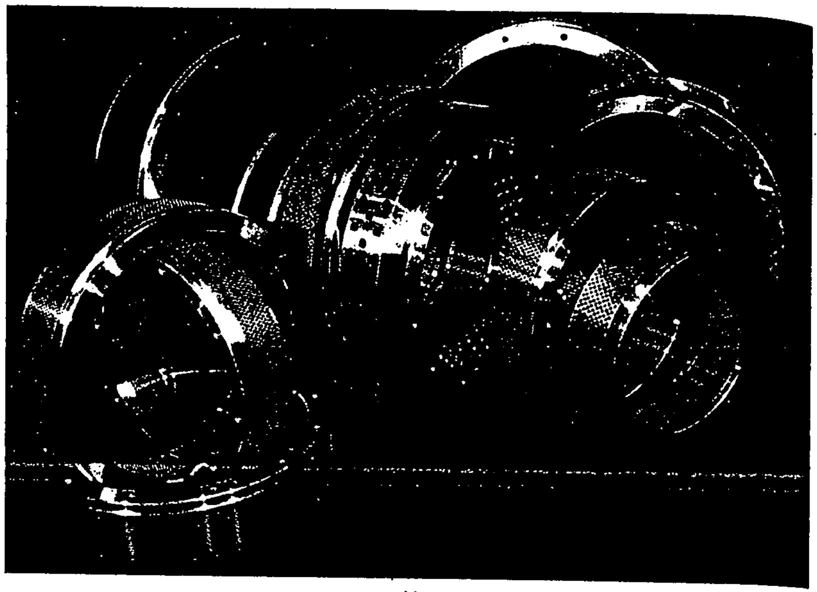
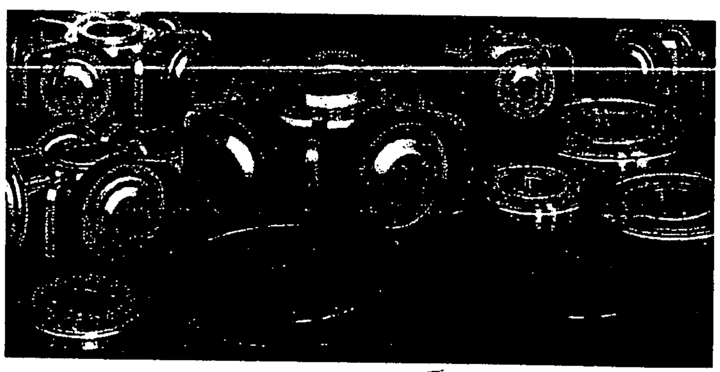


Figure 6.127. (a) Partial cut through a pelleting machine with concave die rings (Andritz Sprout-Bauer, Muncy, PA, USA); (b) photograph of the working parts of a flat die pellet mill (Amandus Kahl Nachf., Reinbek, Germany).



(a)



(b)

Figure 6.128. (*a*) Typical concave pelleting die rings (Andritz Sprout-Bauer, Muncy, PA, USA); (*b*) flat dies and roller assemblies (Amandus Kahl Nachf., Reinbek, Germany).

rollers should be conical if a larger area of the die plate shall be utilized.[15] As with other pressure agglomeration methods, density and strength of pellets can be improved if two

A particular advantage of flat die pellet presses is their applicability for very wet pastes or sticky materials. If the drive mechanism is moved to the top of the machine nothing

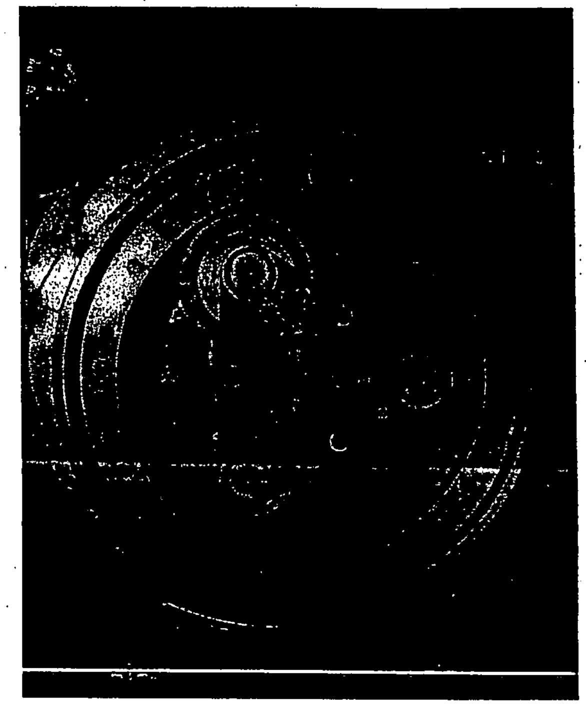
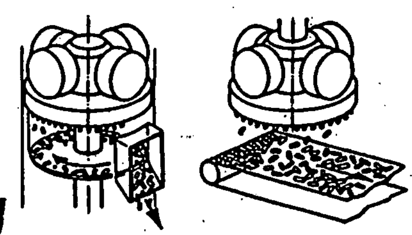


Figure 6.129. Three-roll assembly and feed plows in a concave pelleting machine (Andritz Sprout-Bauer, Muncy, PA, USA).

pellets from the flat die (Fig. 6.130). Pellets from presses with top drive (Fig. 6.130b) either fall as discrete particles onto a fast-moving conveyor where they remain separate entities or discharge directly into a dryer or cooler where they are immediately flushed by air and, therefore, do not stick together.



(a) (b)

Figure 6.130. Diagrams showing two different designs of flat die presses.[11] (*a*) Bottom drive, (*b*) top drive for very wet pastes or sticky materials.

6.3.3 Peripheral Equipment

Conditioning and Product Treatment.

paddle and screw type mixers which can be an integral part of the extruder (see, e.g., Fig. 6.127a) or, in those cases where longer conditioning times are necessary, are separate pieces of equipment.

Figure 6.131 shows three typical mixer conditioners commonly used with pelleting machines.[11] The simple screw type machine (Fig. 6.131a) offers only limited mixing capabilities but is best suited for long, fibrous, and bulky materials. The unit shown in Figure 6.131b combines, in-line, a metering screw and a paddle mixer while the design of Figure 6.131c features a separate metering screw feeding the paddle mixer. Because in the latter arrangement, metering screw and paddle mixer are driven separately, intensive mixing can be achieved at any feed rate. Consequently, relatively large amounts of liquid and/or solid additives can be introduced.

In many cases it is preferential to use steam for heating and moistening; this technique commonly results in higher extrusion rate (capacity), increased die life, decreased power consumption, and improved quality of the extrudate. These characteristics are most reliably obtained if conditioning takes place in separate machines in which residence times of 5 to 30 min can be achieved. Figure 6.132 shows schematically the conditioner of such a system in which material is constantly moved with slowly rotating scrapers and transported from deck to deck while steam is injected and other additives, such as molasses or fat, are incorporated.

Depending on the amount of moisture and/or heat added prior to the extruder, the

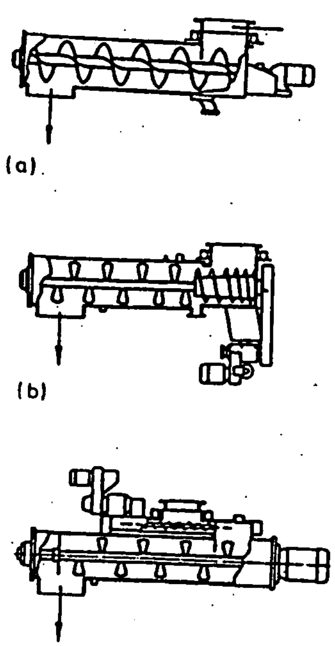


Figure 6.131. Diagrams of three different paddle- and screw-type conditioners.[11]

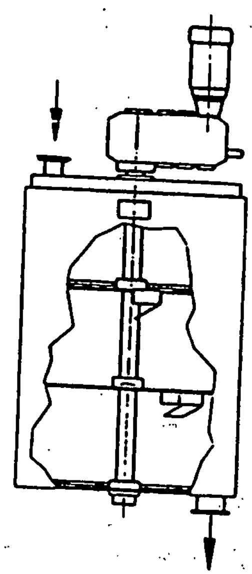


Figure 6.132. Schematic representation of a vertical conditioner with long dwell time.[11]

ct must be dried and/or cooled. Alh sometimes sophisticated equipment is sary for these tasks, simple louvered, verpellet coolers with gravity flow and appli of ambient air for product cooling and g (Fig. 6.133) are commonly used in the al feed industry, which is the single largest cation of pelleting.

izing. As [illegible] ates. Such products are still formable and, therefore, can be further ed in so-called spheronizing equipment to uniform round particles.[16]

Spheronization was developed in the 0/60s,[17] primarily for the pharmaceutical ustry where rounded particles are needed ore uniform coating.

Spheronization begins with wet extrudates ined from one of the previously described uders, preferentially the low-pressure type hines. Because very often small spherical ticles are desired, the extrudates tend to be atively long and thin.

spheronizer consists of a vertical hollow inder (bowl) with a horizontal rotating disc tion plate) located inside (Fig. 6.134). The ghetti-like extrudates are charged onto the tating plate and break almost instantaneously into short segments of uniform length. The friction plate surface has a variety of textures designed for specific purposes. Often, a grid is applied,[16] the pattern of which is related to the desired particle size (Fig. 6.135).

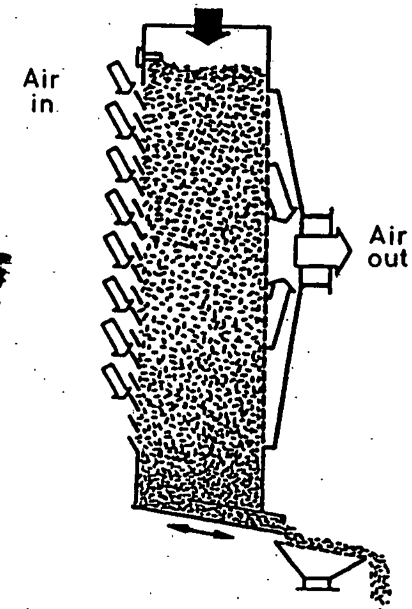
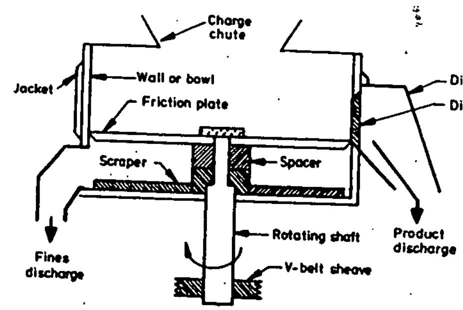
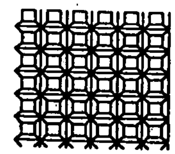


Figure 6.133. Typical design of a louvered, vertical pellet cooler.

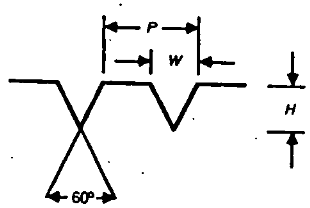




(a)

P	W	H
2	1	1
3	1.8	1.2
5	3	2

(b)



(c)

Figure 6.135. Common grid patterns of the friction plate of a spheronizer.[16] For explanations see text.

For example, a 1-mm granule would be processed on a friction plate with 50% to 100% larger groove openings, that is, 2 mm. The wider groove allows the extrudate to fall into the opening so that the leading edge of the peak will fracture the pellet into pieces with a length-to-diameter ratio of 1.0 to 1.2.

The still plastic pellet segments are being worked by further contact with the friction plate as well as by collisions between particles and with the wall. Mechanical energy is transformed into kinetic energy and the mass of particles rotates in a torus-shaped ring in the apparatus. Continued processing will cause a gradual deformation into spherical shape.

During deformation and further densification excess moisture may migrate to the surface or the mass can exhibit thixotropic behavior. In such cases, a slight dusting by means of a suitable powder dispenser reduces the likelihood [...] cial features may include cooling or heating of the bowl through a jacket and cleaning of the friction plate with brushes.

Spheronization equipment principally operates batchwise. Quasi-continuous operation is possible by means of multiple batches or cascade flow. Either one of these methods used two or more spheronizers. Multiple batch operation, for example, using two spheronizers, is sequenced such that one unit discharges while the other is in the middle of the spheronizing cycle. A reversing belt can be used to alternatively feed each machine. In cascade operation two or more units are linked in series to extend the total residence time. Feed is continuously charged into the first spheronizer and continuously overflows into the next one(s).

6.5.4 High-Pressure Agglomerations

6.5.4.1 Die Pressing[1]

Die presses for compacting powder are the oldest pressure agglomeration machines. They are used by numerous industries for a wide variety of purposes. The largest user is most probably the pharmaceutical industry (see Section 6.5.4.2). However, they are also widely used in the ceramic, powder metal, confectionary, catalyst, and, to an increasing extent, the general chemical industries. The machines can be divided into two main categories: reciprocating or single-stroke machines and rotary machines.

Reciprocating Machines. Reciprocating presses operate with one upper and one lower punch in a single die (see Figure 6.60). They are mainly used for complex shapes where high pressure and/or low outputs are required (less than 100 compressions per minute).

Reciprocating machines can be subdivided into two types: ejection presses and withdrawal presses.

Ejection Presses. Ejection presses are built as very simple hand-operated units and as highly

1000 MN/m^2 pressure and producing compacts with a very high degree of accuracy.

The hand-operating press incorporates basic features common to all ejection presses. The die is mounted in a fixed plate and the upper and lower punches are attached to moving rams. The lower punch descends to allow the die to fill. All the compression is carried out by the upper punch moving toward the stationary lower one. Later, the lower piston ejects the compact upward from the die.

Hand operating machines are very limited in performance. They are only capable of exerting a pressure of 8 to 16 MN/m^2 and the output, obviously, depends on the operator. It is extremely difficult to predict the behavior of particulate matter at high pressure in a rotary press from data obtained by using a hand-operated machine.

A range of mechanical or hydraulic presses has been developed from the hand machine. They vary in the size of compacts that can be produced and the amount of pressure that can be exerted to form the tablet. The smaller machines are used in the pharmaceutical industry for products in which only limited output is required and, to a certain extent, for development work. Larger machines are mainly applied by the powder metal and ceramic industries, but even there, the use is limited in most cases to compacts that feature no change in cross-section, such as washers and short bushings.

The disadvantage of the machines in this category is that they produce a compact that varies considerably in density from top to bottom because the pressure is exerted only by the top punch (see Fig. 6.111). This is not particularly important in the pharmaceutical industry, although in extreme cases it could produce a tablet that disintegrates more rapidly on one side than the other. This disadvantage is of much greater consequence to the ceramic and powder metal industries, where the difference in density will cause uneven shrinkage during sintering. To overcome this "double pressure," that is, the pressure is applied equally to the upper and lower punches.

Withdrawal Presses. Withdrawal presses operate with two cams. The top cam controls the movement of the upper punch and, in turn, the lower cam controls the movement of the die. Whereas the majority of ejection presses are mechanically operated, both mechanical and hydraulic drives are common for the withdrawal type.

In a withdrawal press, compaction and ejection take place with a continuous downward movement of the upper punch and the die (Fig. 6.136).

At the beginning of the pressing cycle, the die is positioned on top of the lower punch to produce the required depth of fill. In fact, the material to be compressed is fed to the die during the return move to avoid the necessity to replace air with the solid feed. The upper punch then descends to compress the material and the die also moves downward during the compression to maintain uniform density in the compressed material. At the end of the pressure stroke, the die continues to move downward until it has been completely removed. During ejection, the compact is supported by the lower punch.

Tooling for this type of press is much more expensive and complex than that required for ejection presses. It consists of a complete die set that is removable from the machine as a complete unit. This has the advantage that the tooling is interchangeable between presses. Further advantages lie mainly in its adaptability to the production of complex components. It is also possible to obtain greater accuracy. Compacts can be made on this type of tooling with dimensional tolerances of less than 4×10^{-5} mm.

In practical terms, apart from the output, effectiveness of mechanical and hydraulic pressure systems is equal. The cycle time of the hydraulic press varies with the stroke. The low-pressure stroke can be made quite fast by using a multistage pump but as the higher

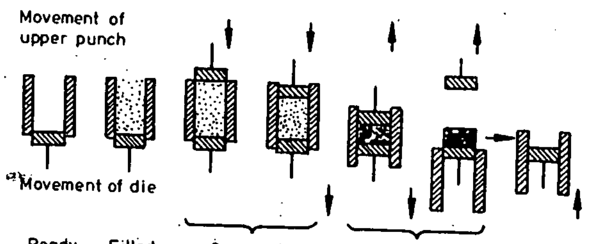


Figure 6.136. Operating phases of a withdrawal press.

becomes progressively slower. The length of the high-pressure stroke depends directly on the thickness of the piece being pressed. In addition, the pumping system of the hydraulic press can rarely achieve a cycle time comparable with the mechanical press when it is used near maximum pressure. Therefore, the use of hydraulic presses is restricted to the ceramic and powder metal industries because of low output and compacts requiring very high pressures. It is also applied in the recycling industry for the reproduction of large, cylindrical compacts from, for example, metal-bearing wastes.

Rotary Machines. Rotary machines were developed to meet the demand for higher outputs of relatively small tablets, primarily in the pharmaceutical industry. Their basic principle of operation is similar to that for hand-operated machines with the exception that the dies are mounted in a rotating table and pass, in turn, under a feed position (Fig. 6.137). The tooling design resembles the one used on the simpler ejection presses whereby the punches are moved by a series of cams. The design of tooling limits the shape of compacts to those that can also be produced on the simpler type of single-stroke machines.

The feed is supplied to the die table by an open frame. The lower punch is pulled down by a cam to the lowest position while the die fills with powder. It then rises up an adjustable ramp, ejecting excess powder from the die. The surplus is scraped off flush with the top of the die table at the highest point of the "weight adjusting ramp," leaving the desired volume of material to be compacted. It is common practice for the lower punch to drop slightly after the surplus material has been scraped off and

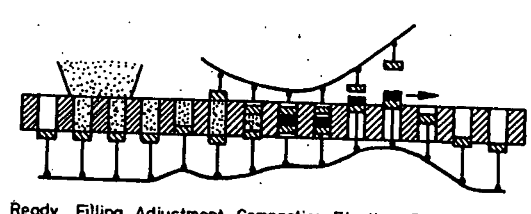


Figure 6.137 Operating schematic of

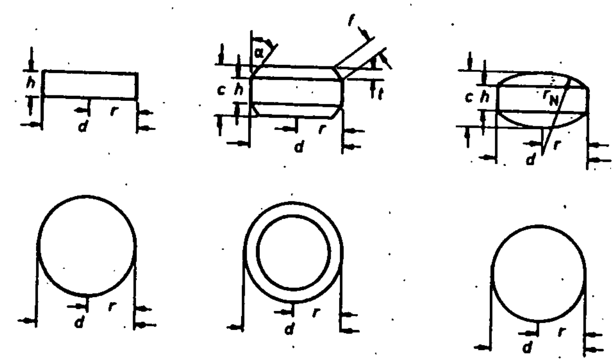
before the upper punch enters the die. This is done to prevent the upper punch displacing material from the die as it enters. The material is compressed by the two punches passing between two rolls, one or both of which are spring loaded. This produces the effect of double pressure. Therefore, the problem of making a compact with uneven density is not very pronounced in rotary machines. Finally, the upper punch is lifted out of the die by a cam and the lower punch travels up another cam to eject the compact from the die.

The simplest type of rotary machines is "single-sided" (one feed location); one tablet is produced from each station (die) per revolution. Therefore, the output of rotary machines depends on the number of stations in the turret (table) and the speed of the turret. It is usually in the region of 300 to 800 tablets per minute. A further increase in output is possible by using a "double-sided" machine. In this case the stations are filled twice on opposite sides of the rotating table; two compressions are carried out in each die per revolution of the turret. Outputs of up to 3000 tablets per minute can be obtained from the "double-sided" machine. Still further capacity increases can be obtained by dual or multiple tooling (two or more dies) per station.

Special Design Features of Die Presses. (Many of the schematic drawings used in this section are reproduced from the "Powder Metallurgy Equipment Manual"[18] with permission of the Metal Powder Industries Federation. Special die presses for the pharmaceutical industry are described in Section 6.5.4.2).

Shapes. The original and still most common shape of die pressed agglomerates is a more or less cylindrical "tablet' (Fig. 6.138).[19] Included in this description are flat, faceted, and crowned compacts. For these shapes, simple die and punch configurations are applicable. Structured shapes can be necessary in Powder Metallury (P/M) where a classification of I through IV characterizes the complexity of part design.[18]

One-level, relatively thin tablets or parts with any contour (Class I of PM, Fig. 6.139) can be pressed with a single punch and force may be applied from one side. The maximum dimension A (Fig. 6.139) depends on the particulate feed and the shape of the compact. Thicker parts (Class II of P/M, Fig. 6.140), while still requiring only simple tooling, must be pressed from two directions. Holes are obtained by the installation of mandrels or core rods.



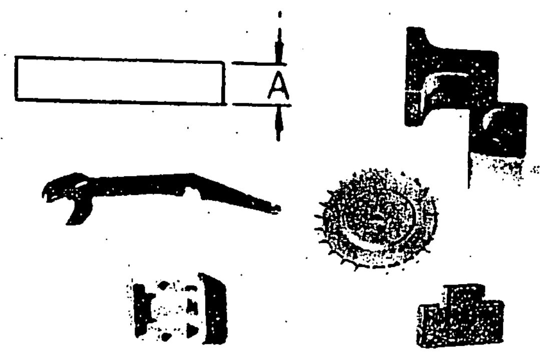
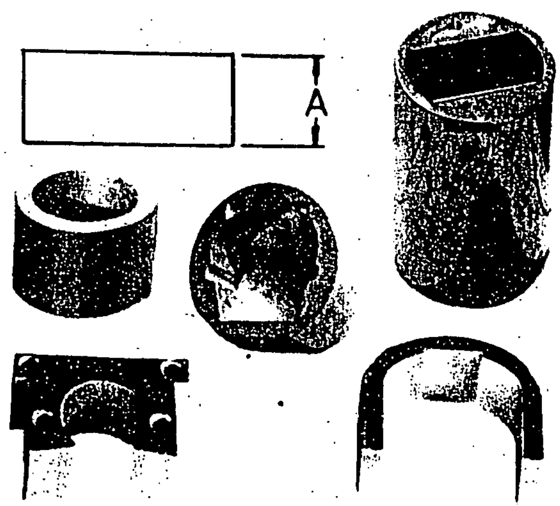


Figure 6.139. P/M classification: Class I parts.[18]

Because, owing to interparticle friction there is little or no hydrodynamic flow of particulate solids during compaction, each level of more complicated parts must be supported with a separate punch or die member to maintain reasonably uniform density throughout the green pressed part (Class III and IV of P/M, Figs. 6.141 and 6.142).

Drives. The above product shapes are usually made in mechanically operated die presses. Advantages of mechanical presses are: high production rates, low power requirements, and a large range of applicable pressing forces. The most common mechanical drives are: eccentric or crank, toggle, cam, and rotary arrangements.



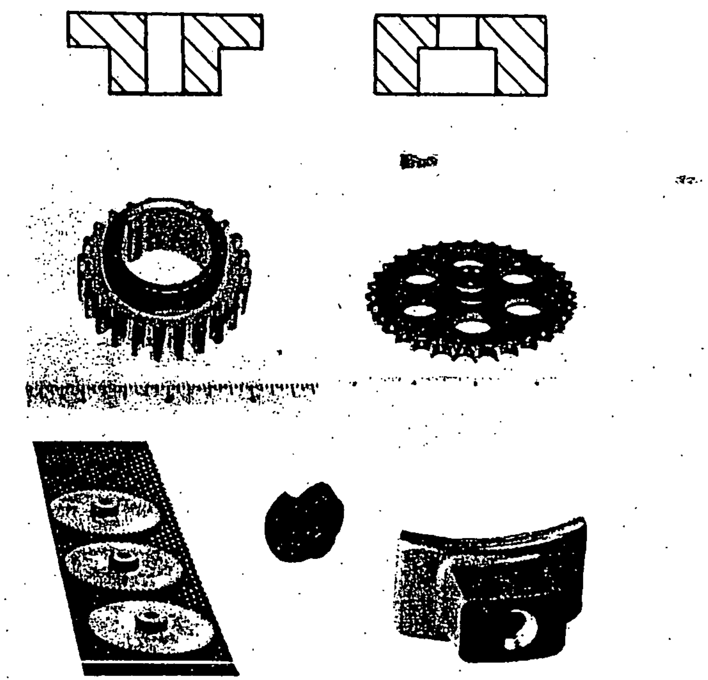


Figure 6.141. P/M classification: Class III parts.[18]

Figure 6.143 represents eccentric or crank type drives which convert rotary motion to linear, reciprocating movement. The mechanisms feature small final rate of pressing speed (approaching bottom dead center) and high loading with low torque at maximum compression (at bottom dead center). The stroke can be adjusted on the eccentric cam or "Pitman" link. Normally, this method is used when force is applied from only one side and, typically, it drives the top punch.

Another common drives mechanism is the toggle (or knuckle) type (Fig. 6.144). Actuation is normally accomplished by eccentric or crank arrangements that alternatively straighten and bend a jointed arm or lever. If one end of the lever is fixed, the other—if guided properly—will produce a linear movement whose stroke can be adjusted as mentioned previously. Final pressure will be even higher and pressing speed near the end of compression is minimal.

Figure 6.145 depicts schematically the cam drive. Cam and lever arrangements are used to convert rotary motion to linear movement. Pressing speed, timing, and motion are adjustable by changing the contours of the cams or cam inserts.

The cam drive is mostly used for rotary die presses which feature a series of punches and dies arranged in a common, rotating, tool holding table (turret) (see also Section 6.5.4.2 and Fig. 6.163). The stationary axis around which the turret rotates provides a fixed reference point for mounting the press cams and

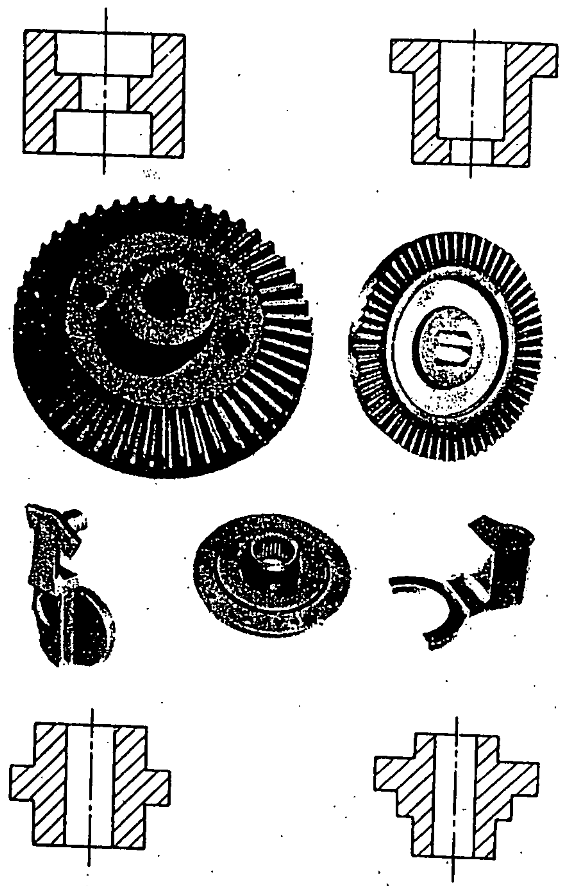


Figure 6.142. P/M classification: Class IV parts.[18]

The disadvantage of all mechanical punch drives is that, while compression speed becomes smaller as the eccentric connection of the rotating drive member approaches dead center and cam drives may follow curves that allow a certain "dwell-time" at maximum compression, compaction takes place very quickly with a sudden release of force after reaching the maximum. This is a particular problem if the material to be compacted ...

erties. Such products reach sufficient permanent (plastic) deformation and strength only after remaining under pressure for some time. Premature pressure release results in excessive elastic spring-back which may destroy the structural integrity of the compact and result in well-known failure modes (e.g., capping, lamination, etc.) indicating "overpressing."

The only reliable way to overcome this

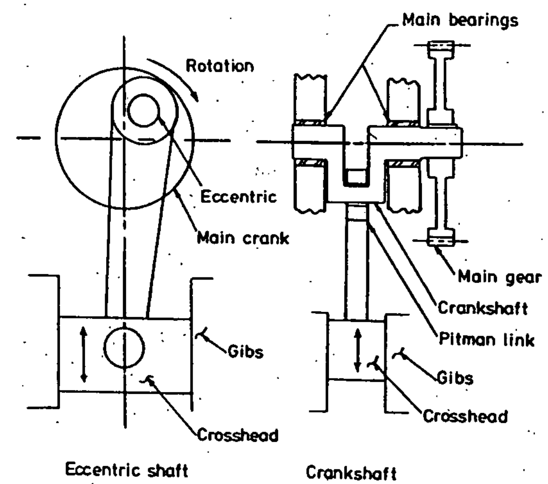
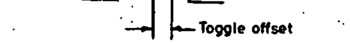


Figure 6.143. Eccentric or crank drive arrangement.[18]

actuation of the punch(es) (Fig. 6.146). The timing of the punch strokes as well as the rate of increasing or decreasing pressure and the "dwell-time" can be easily adjusted. In addition, hydraulic presses typically feature overload protection by means of gas filled accumulators and allow the densification of larger amounts of feed even with low initial bulk density. Because there is no physical limit to the length of the stroke, densification ratios can be very high; and, since pressure rise can be slow, final pressure high, and "dwell-time" adjustable without limiting constraints (other than capacity), elastic materials, such as organic refuse or other organic materials and, for example, steel turnings can be successfully compacted. Figure 6.147 is the sketch of a large, hydraulic, horizontally oriented high-pressure press.

More conventional presses feature vertical design (Fig. 6.148). They can be highly automated and, with multiple tooling, producing several compacts per stroke, as well as auto-



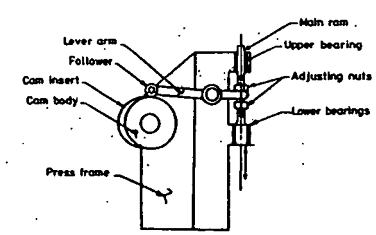


Figure 6.145. Schematic representation of the cam

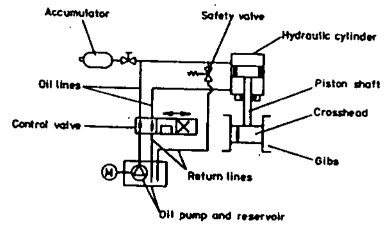


Figure 6.146. Schematic representation of a hydraulically driven press.

matic feeding and product handling systems, can have considerable capacities. Typical applications are in the refractory industry for making brick. But many other uses are conceivable as demonstrated in Figure 6.149, which shows a selection of parts.

Press Feeders. To obtain parts with high accuracy of volume and density it is necessary to employ automatic powder feeders. The design of such mechanisms is simpler for the otherwise more complicated rotary presses then for the much less complex machines with fixed tool carrier table. Rotary presses employ a stationary filling shoe (see Section 6.5.4.2, Fig. 6.162); because of the high rotational speed of the turret the feed must be free flowing and, therefore, is often preagglomerated (granulated). To further improve feeding and guarantee uniform filling at high rotating speeds, "power feeders" are employed. Their design is such that they can be easily removed and opened for cleaning.

Feeders for presses with stationary tool holders can be divided into direct shuttle, metered shuttle, and arc type feeders.[18] The direct shuttle feeder (Fig. 6.150) may also be used on moving die tables. It provides a straight in-line reciprocating action over the die with the feed shoe connected directly to the supply hopper. The motion of the metered shuttle feeder (Fig. 6.151) is the same as that of the in-line system (Fig. 6.150) and may also be applied on moving die tables. It does not have a direct connection with the supply hopper. At fill, it moves with a metered amount of material from a position under the hopper to the die cavity. This system supplies the same amount of material on each press stroke. The arc type feeder (Fig. 6.152) is normally applied only on mechanical presses with a stationary table. It uses a pivoting action of the feed shoe over the die area.

Control of the lower punch and the feed shoe is typically such that material is transported to the die area when the punch is still in or near the ejection (highest) position. This avoids the cavity filling with air which must be replaced by feed and finally expelled during compaction. Particularly with high-speed

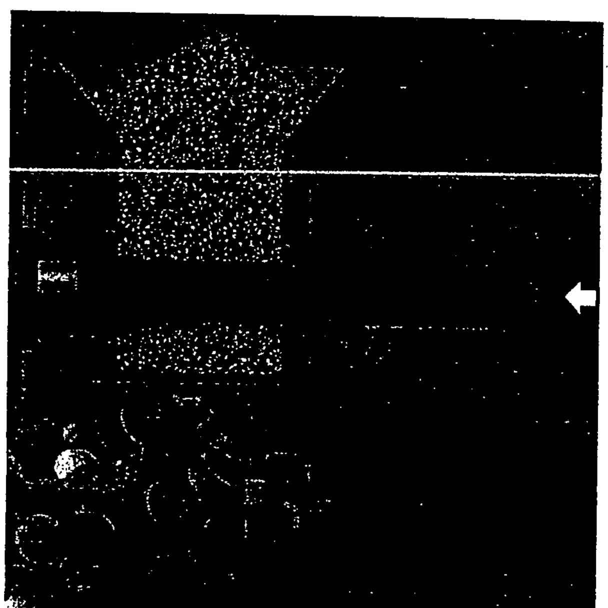


Figure 6.147. Diagram showing the principle design of a large hydraulic press with horizontal punch movement (Lindemann, Düsseldorf, Germany).

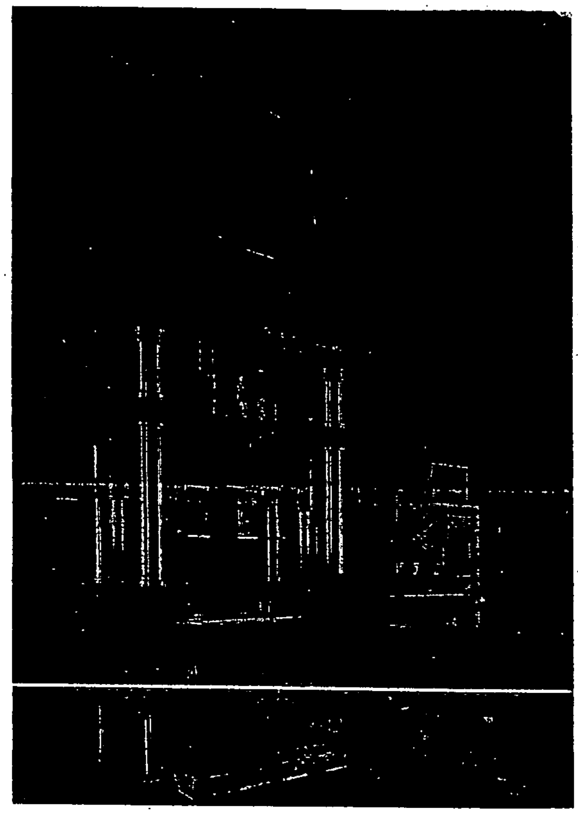


Figure 6.148. Typical vertical hydraulic press for the manufacture of refractory brick (Horn, Worms, Germany).

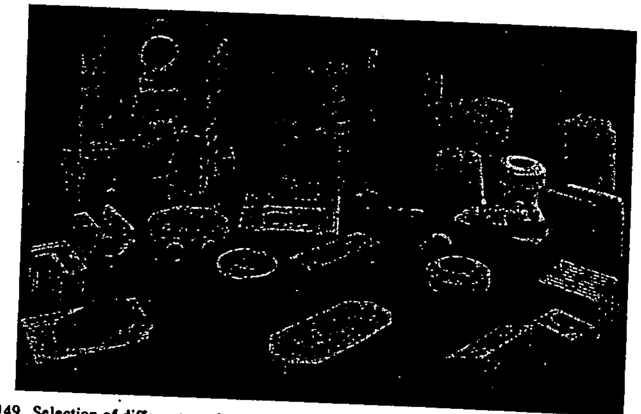


Figure 6.149. Selection of different products made with vertical hydraulic presses (Horn, Worms, Germany).

presses, sufficient deaeration may pose a major problem and compressed pockets of air can be an important cause of tablet failure (e.g., capping).

Tooling Design. Since particulate solids do not flow under pressure, friction within the mass and on the tool walls absorbs part of the force applied by the punch(es). The "neutral axis" is the low-density zone approximately perpendicular to the direction of pressing. Control of the location of this zone in the compacted part is often important (e.g., to avoid distortion of P/M parts during sintering) and is achieved by the relative tooling motions. Under pressure, particulate matter will also not flow from one part level to another. Therefore, when parts of more than one level are pressed,

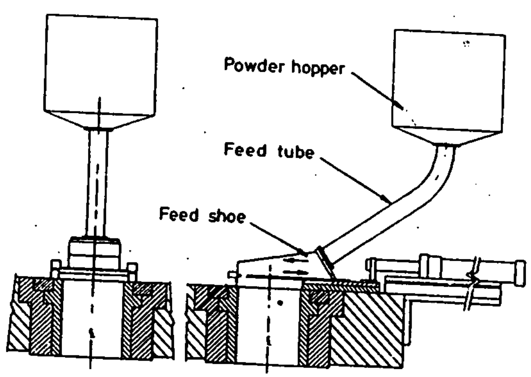


Figure 6.150. Schematic representation of the direct shuttle feeder.[18]

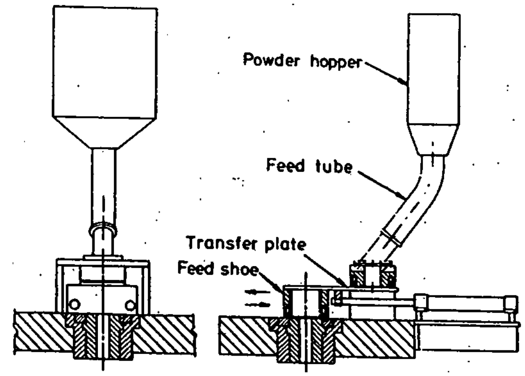


Figure 6.151. Diagram of the metered shuttle feeder.[18]

separate pressing forces must be applied simultaneously for each level. As a result, there will be a neutral axis for each part level (Fig. 6.153).

Figure 6.154 demonstrates how the location of the neutral axis of a simple, one-level part can be controlled in a die press with upper punch pressing and controlled withdrawal die (see also below and Fig. 6.159).

As far as variety of applications, complexity of shapes, and accuracy of parts are concerned, die pressing is the most versatile agglomeration method. To achieve this versatility, the basic principle of die pressing is often modified. The most important methods, reflecting the significance of the technology, are reviewed in the following.

Single-Motion Pressing

This is the simplest method and is usually limited to compacting relatively thin parts with or without through holes obtained by the installation of core rods. Only one part of the tooling is moved during compression.

Figure 6.155 depicts schematically the three stages of upper punch pressing. Other single-motion pressing designs are sketched in Figure 6.156. During sliding anvil pressing (Fig. 6.156a) the lower ... plishes filling, compaction, and ejection. Normally, powder feed, anvil, and pick-up are three separate components brought in place by a "positioner." Figure 6.156b shows the "Pentronix unitized anvil" in which all three functions are combined into one assemblage which is always in contact with the die plate. Powder spillage and blow-out are reduced to practically zero, making this design ideal for, for example, the processing of toxic materials. In anvil withdrawal pressing (Fig. 6.156c) the lower punch remains stationary while the die table is moved into positions for filling, compaction (with anvil in place), and ejection.

Double-Motion Die Pressing

This method will produce parts with more uniform density. Double-motion pressing provides force to the particulate mass to be compacted simultaneously from top and bottom through movement of two parts of the tooling, for example, the upper and lower punches (Fig. 6.157). A similar effect can be obtained by upper punch pressing with floating die (Fig. 6.158) whereby the die table moves if the frictional forces overcome the supporting or counterbalancing force holding the die. This die travel has the same effect during com-

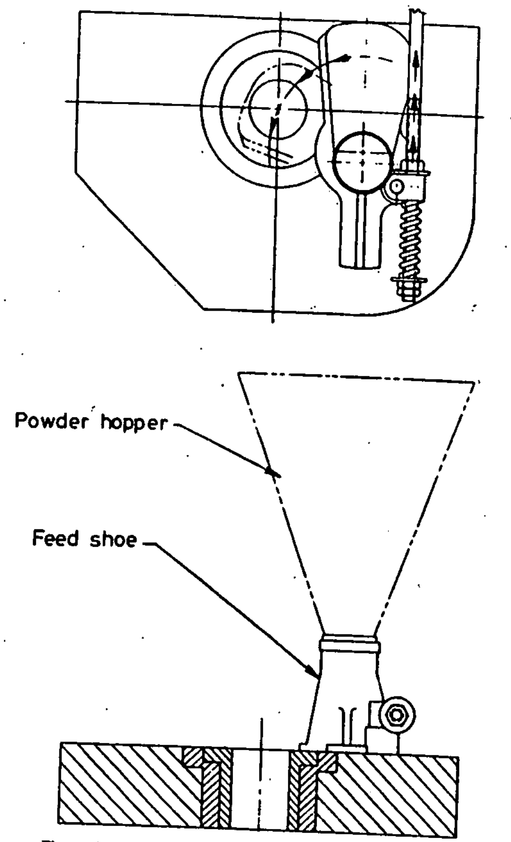


Figure 6.152. Diagram depicting the arc-type feeder.[18]

be accomplished by movement of either the lower punch (Fig. 6.158a) or the table (Fig. 6.158b, upper punch pressing, lower fixed punch, floating withdrawal die). Potential disadvantages of this system are that compacted

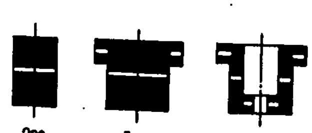


Figure 6.153. "Neutral axes" in single- and multilevel

parts may have density variations that are determined by the size of the supporting force of the die and that the neutral axis may not be located in the center of the part.

In upper punch pressing with controlled withdrawal die (Fig. 6.159) adjustment of timing of die travel provides positive control over the position of the part's neutral axis (see also Fig. 6.154).

Multiple-Action Pressing

Multiple-action pressing systems are those that

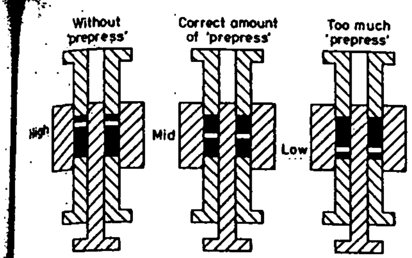


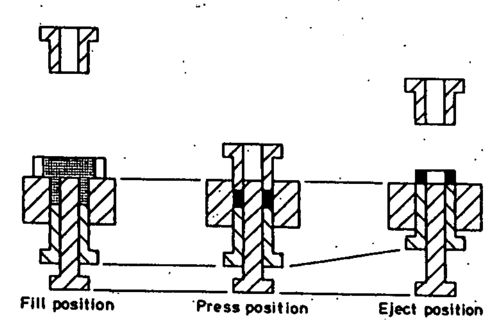
Figure 6.154. Possibilities to influence the "neutral axis" position in upper punch pressing with controlled withdrawal die.[18]

parts with a separate punch or tool member. Such tooling is used to minimize density gradients in complex compacts. Therefore, all of these more sophisticated machines also use one or the other method of double-motion pressing as demonstrated in Fig. 6.160. With descriptions the figures are self explanatory.

In the foregoing, the need to minimize density variations was mentioned several times. Excessive density gradients may cause destruction of compacts (capping, laminating, cracking, etc.) and deterioration of parts during finishing (firing, sintering, etc.). In addition to double-motion pressing and multiple-action tooling, it is sometimes necessary to decrease friction by the addition of lubricants. Because in most cases lubricants are impurities and costly it is desirable to keep their amount as low as possible. Lubricants can reduce interparticle and/or die wall and tooling friction. In most cases, however, the lubrication is required only on the die walls and tooling. In fact, if lubricants are blended into the mixture to be compressed, the normally hydrophobic additives may reduce product quality. Therefore, new developments are directed toward the lubrication of only the tool surfaces.

Tooling design, tolerance, and finish are of utmost importance for die pressing and the quality of compacted parts. The die holder (table, turret, etc.) normally has larger holes into which die inserts are mounted. Whereas for simple, cylindrical contours sleeves can be clamped or shrunk into the openings, designs and mounting of noncylindrical die configurations require considerable know-how and skill. The problem is aggravated by the need to produce dies from abrasion-resistance material (e.g., carbides) and to provide tight tolerances with high-quality surface finish. Often, dies must be made up of different parts as shown, for example, in Fig. 6.161.

For improved deaeration and release of the compacted part, die walls and core rods are often slightly tapered. However, clearances must be small enough to retain the particulate solids in the compression chamber. Die cavities and core rods must has a high-quality surface finish (polished, lapped, etc.) and strong supports must be provided to avoid distortion under pressure. In multiple tooling arrangements, some punches must also partially serve as a die. In such a case, the punch



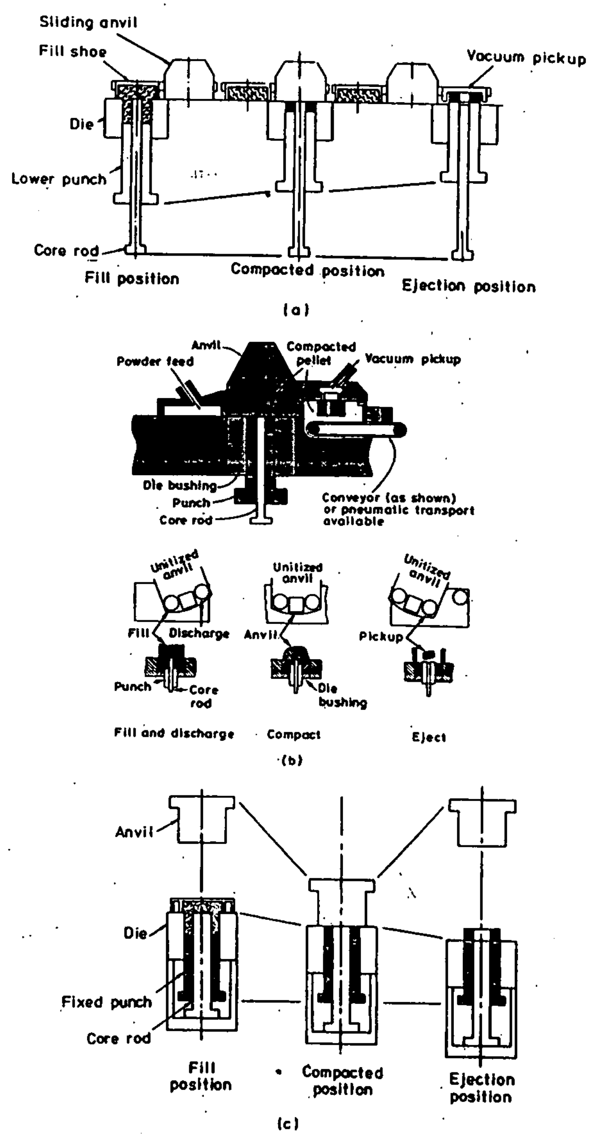


Figure 6.156. Different sketches representing anvil pressing. (*a*) Sliding anvil,[18] (*b*) unitized anvil (Pentronix, Lincoln Park, MI, USA), (*c*) anvil withdrawal pressing.[18]

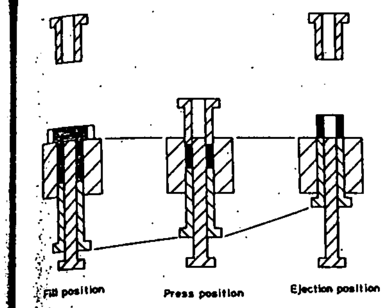


Figure 6.157. Double-motion pressing.[18]

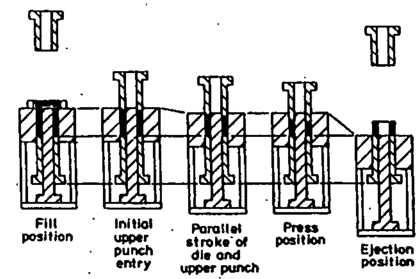
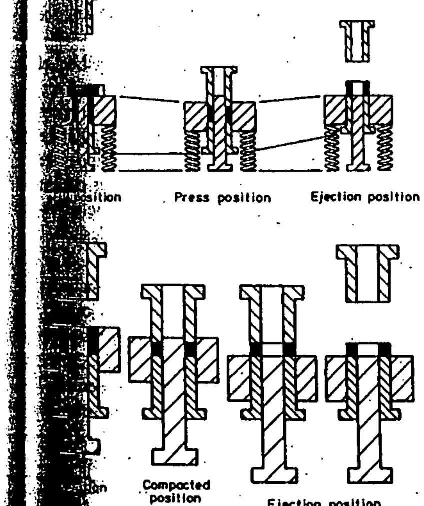


Figure 6.159. Upper punch pressing, controlled withdrawal die.[18]

must be backed up for the full length of the compact to provide rigidity.

In addition to these basic designs and general requirements there are large numbers of supplemental machine and tooling options. Requirements for nonstandard equipment or press characteristics must be determined for each particular application and discussed with the machine manufacturer. As usual in the field of agglomeration, suppliers of die presses maintain well-equipped technical centers in which special requirements can be tested and machine modifications are developed as necessary.



...158. Sketches depicting presses with floating ... explanation see text.

Today, the main thrust in new developments for die presses in in the area of machine data measurement and control.[20] Techniques have become available to accurately measure the parameters during a press cycle that may last only a few hundred milliseconds. Based on such information, production machines can be programmed and automatically controlled.

6.5.4.2 *Tabletting in the Pharmaceutical Industry*[21]

Compressed tablets are the most common pharmaceutical dosage form. The reasons for this are: (1) they are convenient, compact, easy to carry and ship, and (2) they are usually more chemically stable than other dosage forms, since most drugs decompose by hydrolysis.

This was the first paragraph of Chapter 7, Part 3 of the first edition of this book in which Carstensen[21] described in much detail "machines, manufacturing procedures, formulation parameters, and basic principles on which formulation principles are based." He also included an extensive list of machine specifications, "since such a compilation has been

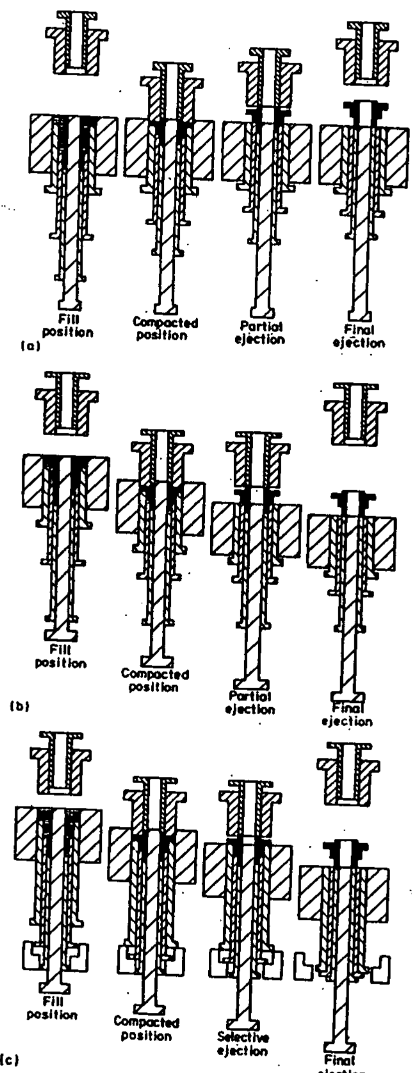


Figure 6.160. Schematic representation of three multiple-action tooling systems.[18] (*a*) Upper and lower punches pressing, die stationary; (*b*) upper punches pressing, floating withdrawal die; (*c*) upper punches pressing, controlled withdrawal of die and lower punches.

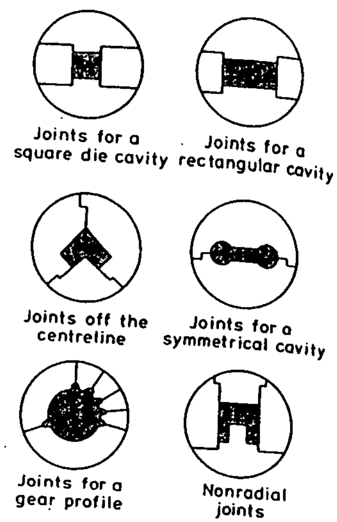


Figure 6.161. Six examples of die inserts with preferred location of joints for noncylindrical cross-sections of parts.[18] The shaded area is the die cavity.

deemed useful for both the reader and the practitioner."

Because this topic is very specialized but of considerable interest to a large industrial segment using pressure agglomeration, a much shortened version will follow. In particular the "extensive lists of machine specifications" are not given because they are no longer current and can be obtained readily from the manufacturers of tabletting equipment if desired.

Tablet Machines. The first tablet machines were introduced in the nineteenth century, and have by now been developed into sophisticated, high-precision tools.

They may be either single-punch (eccentric) machines (Fig. 6.162) or rotary presses (Fig. 6.163, see also Fig. 6.137).

In the *eccentric machine*, powder flows from the shoe into the die in position 1. The shoe then swings away, and the upper punch is lowered to compress and powder (position 2). Both punches then are raised (position 3), lifting the tablet out of the die, and the hopper then comes back into its original position (and knocks the ejected tablet onto the discharge chute). The powder level in the shoe is maintained by gravity feed from the hopper.

The fill weight can be adjusted by the (low) position of the lower punch. The lower it is, the higher the fill weight. The fill weight is also a function of the apparent density and the flow rate of the powder. The compression pressure (and hence the tablet hardness and porosity) can be adjusted by the (low) position of the upper punch.

In a *rotary press* there is a series of dies positioned circularly on a die table (Fig. 6.163a). The upper and lower punches glide on cams (Figs. 6.163b, c, d). An evoluted picture is shown in Figure 6.163b. The filling takes place between points *A* and *B*, that is, under the feed frame. This in turn is fed by the hopper. The powder is leveled (scraped) at point *B*, so that the fill is a function of the level of the lower punch at this point. As the table rotates (goes from right to left in Figure 6.163b), the die passes the feed frame, and the lower punch drops a small amount. With the pressure wheels, the upper punch is brought down and the lower punch raised to form the tablet. Both are then raised (by the cam contour), and the tablet is ejected. Point *A'* corresponds to *A* (the back end of the feed frame which serves as an ejection bar for the tablet). It is obvious from the drawing that tablet weight can be adjusted by screw *E*, ejection by screw *F* (where the ejected tablet must be flush with the table) and compression pressure by the relative position of the pressure wheels.

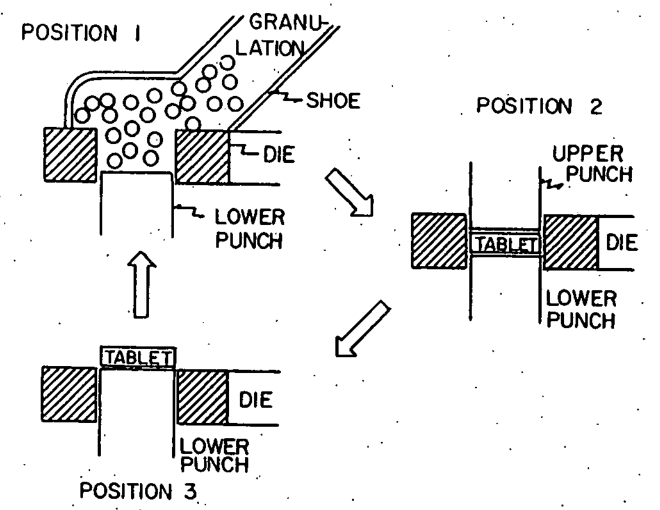


Figure 6.162. Steps in the formation of a tablet on a single punch (eccentric) machine.

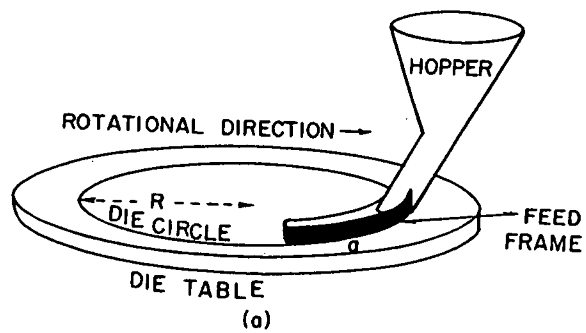
In the simplest case of a rotary machine there is one hopper and a certain number of "stations" (as few as four) on the die table. In other words, one rotation produces the number of tablets given by the number of dies (and punch sets) on the machine.

Expulsion of entrapped air from a granulation or (particularly) a powder mix is important since it reduces lamination and capping of the produced tablets. High-speed machines are equipped with a precompression feature. Solids for tableting are of three types: (1) noncompressible powders, (2) compressible powders

possessing poor flow, (3) compressible powders possessing good flow. Noncompressible powders are either wet granulated (which adds a binder, making them compressible) or (if they are of sufficiently low dosage level) mixed with a powder excipient of type (3), so that the mixture is compressible and free flowing.[22]

When powders are granulated, flow characteristics are usually superior to those of naturally free-flowing powders; hence direct compression powders (i.e., mixtures of type 3) are usually aided in the filling step of artificial means, the so-called forced feeders. Flow in the hopper can be of concern also (and if not uniform will cause inconsistent tablet weights).

Powders of type (2) (especially if moisture sensitive) will form tablets, but because of inconsistent flow they cannot be compressed



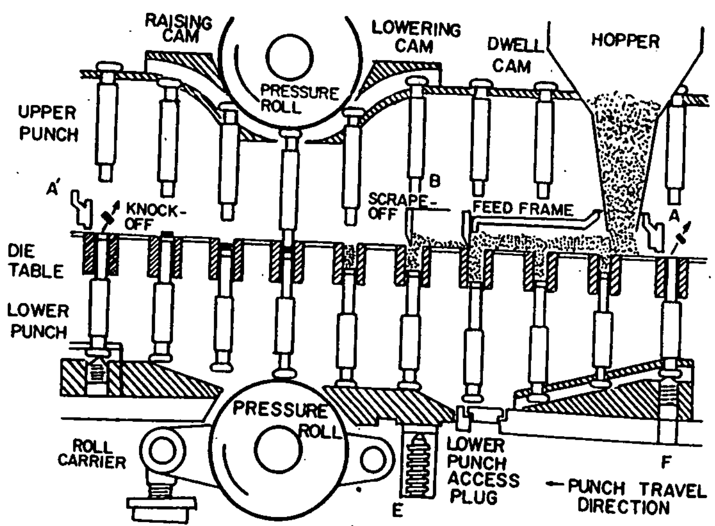
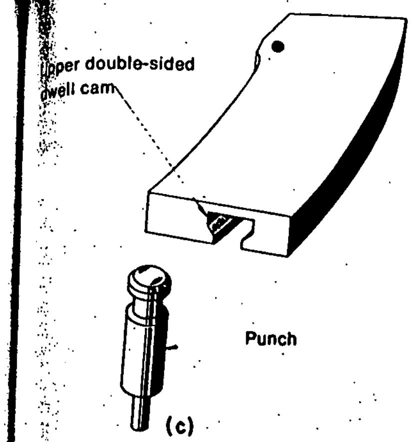


Figure 6.163. (*a*) Schematic of a rotary machine. (*b*) Path of punches during tableting on a rotary machine in evoluted presentation.[21]



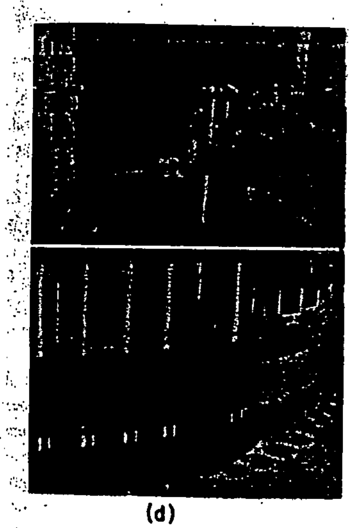


Figure 6.163. (*c*) Double-sided upper cam.[21] (*d*) Photograph of punch being installed on a tableting press.[21]

directly to produce tablets with uniform tablet weights. The flow in these cases is often improved by particle-size enlargement effected by first making large tablets (slugs, boluses) on a heavy-duty machine or compacting the mixture in sheets with roller presses. These slugs or sheets are broken up by milling through a suitable screen, to from fragments of a larger particle size than the parent powders. Hence flow is better, and tablets can be produced that have satisfactory weight variation.

In should be noted that a fair amount of development of tablet formulas is done at a stage where only small amounts of drug are available (the so-called stages I and II in the clinical progression of drug development), and that scale-up difficulties into high-speed equipment can be anticipated. Because of government requirements that eventual production formulas be identical to those tested in the clinic, severe pilot problems always exist in the pharmaceutical industry.

In many cases there are incompatibilities[23] among drugs, and such solids are kept apart from one another by special means, notably by double- or triple-layer tablets or by compression-coated tablets (tablet within a tablet). In the triple-layer tablet, compression takes place in several stages, requiring a special press. There are three hoppers, 120° apart on the die table. Filling takes place in three steps. In the first stage, the low position of the lower punch dictates the fill weight (of the first layer); in the next stage the "bottom" of the die is the top layer of the first filled powder; and in the last stage, the surface of the second layer is the "bottom" of the die. Intermediate "tamping" is possible, and this, for instance, improves the precision of the fill of each layer.

In the case of compression coating, a tablet is first manufactured on one press (which constitutes one half of the total press assembly), and then transferred into the half-filled larger die, with the "outer" granulation on the other half of the machine. This is then filled to the top and compressed. In both compression coating and multiple-layer tablets, the intergranulation and layer bonding and the amount of moisture are exceedingly important parameters.

In a triple-layer table, the precision of fill is less than in a conventional tablet, as far as the

individual layers are concerned. Defects are primarily (1) insufficient interlayer bonding, giving rise to separation of layers, (2) unevenness of the layers (which can be seen directly if multicolor schemes are employed). In the compression-coated tablet the defects are (1) missing core, (2) poorly centered ore, which can be seen from the "outside" of the tablet, and (3) splitting caused by inadequate bonding in the outer layer. The formulation of these types of products is difficult.

Tablet Formulations. To formulate a tablet one must first know the desired size as well as shape and approximate thickness. In this manner one may estimate the approximate weight. The sum of all ingredients is, of course, the tablet weight, and estimates are then made of the required amounts of necessary ingredients. The amount is then brought to the desired weight (q.s.) with filler. A list of ingredients and approximate concentration ranges is shown below:

DRUG	EXAMPLE	RANGE
Disintegrant	Cornstarch	0–8%
Lubricant	Magnesium stearate	0–2%
Glidant	Talc	0–1%
Binder	Cornstarch	0–5%
Filler	Lactose	q.s.

Except when placebo tablets are made, the drug is present, and in an amount dictated by its nature. When a tablet is administered to a patient, it must *disintegrate* in the gastric (and intestinal) fluids. On contact with biological fluids, swelling substances such as starch, certain resins, alginic acid, and modified polyvinylpyrrolidone will expand sufficiently to "blow" apart the tablet.

When a tablet is compressed in the die, a residual force exists against the die wall (Fig. 6.164). This force P is perpendicular to the ejection force E, exerted by the lower punch during the ejection phase of the tabletting. The two forces are related by the frictional

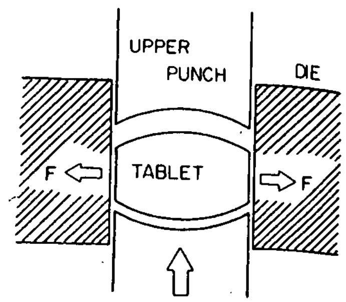


Figure 6.164. Residual die wall force F and ejection force E.

coefficient F. The function of the lubricant is to reduce the value of F. Improperly lubricated formulations will, in milder cases, give rise to tablets that are prone to cap (or that actually do cap), that is, the crown separates from the rest of the tablet. Hairline cracks in the walls of the tablet are usually indicative of this condition. In more severe cases the formulation will "bind up" in the die, and the tablet machine will stop operating. There are formulation reasons for capping as well; for instance, a too large quantity of fines will give rise to capping. The actual capping often occurs as the tablet is being ejected (i.e., actually outside the die), because at this point the tablet expands. Lubrication, machine speed, and reduction of fines are usually the remedies employed in the case of "capped tablets."

To obtain a good tablet the powder of granulation must flow wall. *Glidants* are sometimes added to improve flow, but most frequently flow is controlled by particle size and surface. The property affected by poor flow is the consistency of the tablet weight. The United States Pharmacopeia XVI states the following requirements for weight: of 20 individually weighted tablets only two may differ from the mean by more then the stated percentage, and the tablet may differ by more than twice the stated percentage:

Tablets weighing 13 mg or less	15%
Tablets between 13 and 130 mg	10%
Tablets between 130 and 324 mg	7.5%
Tablets more than 324 mg	5%

[redacted]re added to tablet formulations to produce granules or powders that will bind together to make a good compact in the tablet [redacted] The pastes used in wet granulation[23] are mostly: Cornstarch paste (0 to 10%), sucrose (usually added dry, water being the granulating liquid), povidone (polyvinylpyrrolidone) (10% alcoholic solution), acacia (10% aqueous solution), and gelatin (5 to 13% aqueous solution).

[redacted]are usually sugars, sugar alcohols, or inorganic substances. Lactose, dicalcium phosphate, sucrose, and mannitol are common tablet fillers. All nondrug substances in a tablet are denoted excipients.

Factors Affecting Flow and Compression. Flow rates of powders affect tableting in two ways: the flow from the hopper to the feed frame must be adequate, the flow from the feed frame to the die must be adequate.

Powder flow is a function of

1. Particle size
2. Particle shape
3. Roughness of surface
4. The chemical nature of the compound (cohesion)
5. Moisture.

In general, flow versus *particle diameter* is a parabolic function, such as shown in Figure 6.165. The maximum (d_m, W_m), where d and W are diameter and flow rate, respectively, occurs at fairly large diameters (400 to 1000 μm), so that flow problems associated with fineness and cohesiveness of powders can usually be solved by particle enlargement. The general methods employed are either wet or dry granulation or slugging.

The effect of the *particle shape* has been described by Ridgway and Rupp.[25] They define a quantity for describing particle shape (shape factor) in the following fashion: If d denotes the projected mean diameter of the particle, it is possible to express the surface A

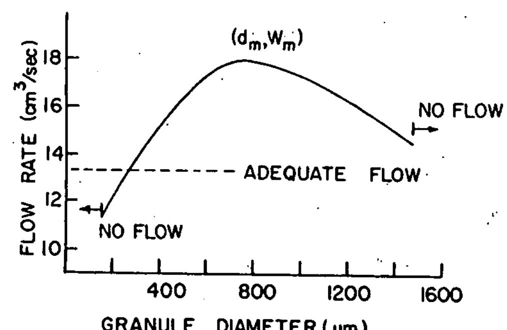


Figure 6.165. Flow rates as a function of particle diameter.[24]

and the volume V of the particle as $A = q_1 d^2$ and $V = q_2 d^3$, and the shape factor is then $G = q_1/q_2$. In general the effect of the shape factor on flow amounts to a 20% drop in flow rate with a doubling (from, e.g., 7.5 to 15) of shape factor.

The effect of orifice diameter on flow is described by the Brown and Richards Equation[26]

$$(4W/\pi\rho g)^{0.4} = mD + C \qquad (6.52)$$

The effect of the *addition of fines* to a monodisperse powder has been described for instance by Danish and Parrott.[27] The general effect of this step is shown in Figure 6.166. The amount of material that can be filled into a tablet die is the apparent density ρ' (g/cm³) multiplied by the volume V (cc) of the die cavity. If the contact time between the die and the feed frame of length a (cm) is t (seconds), then on a die table of radius R (cm) and rotational speed Ω (rotations per second),

$$t = \frac{a}{\Omega 2\pi R} \qquad (6.53)$$

In general, as long as the flow rate has a value over a critical value W' given by:

$$W' = V\rho'/t = D\Omega 2\pi/R/a \qquad (6.54)$$

the fill weight will be D (g). However, for values of $W < W'$, this is not the case, and here the tablet becomes a function of flow rate:

$$D = Wa/(\Omega 2\pi R) \qquad (6.55)$$

These relations are shown in Figure 6.167. There is no sharp break between the two linear portions predicted by Eqs. (6.54) and (6.55), and on high-speed machines, the situation is frequently in the transitional region (the curve in Fig. 6.167).

The thickness h (cm) and the hardness H (kg) of a tablet are functions of the pressure P (Pascals) applied in the formation of the tablet. This, of course, is a function of the relative distance between the two punches at their closest point of approach. The thickness h follows the Fell–Newton law:[28]

$$\ln \frac{h - h_\infty}{h_0 - h_\infty} = -k(P - P_i) \qquad (6.56)$$

and this relation is shown (in linear fashion) in Figure 6.168. h_∞ is a function of the true density of the tablet ρ (g/cm³), in that the (nonporous) mass of the compact is given by:

$$D = h_\infty \pi (D/4)^2 \rho \qquad (6.57)$$

h_0 is given by the apparent density (ρ') in a similar expression:

$$D = h_0 \pi (D/4)^2 \rho' \qquad (6.58)$$

Equation (6.56) applies only to the last steps of compaction and hence P_i somehow relates

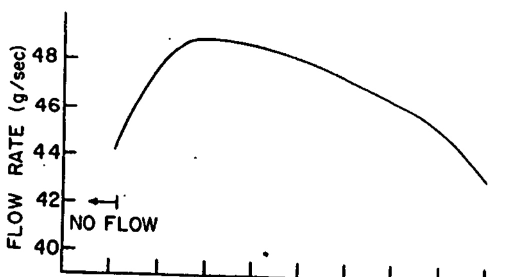
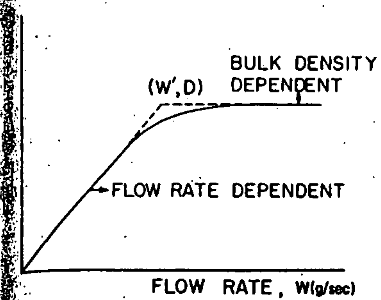


Figure 6.166. Fow rate as a function of percent fines in a granulation.[27]



...re 6.167. Fill weight as a function of flow rate of a ...ulation or powder. D is dose and W' is the critical ...rate.

...the elastic limit beyond which deformation ...longer gives rise to the same shape or size of the particle when the pressure is released. The rate at which a powder or a granulation ...consolidate may be critical when high-...ed machines are used, and therefore, con...dation rates play a part in compression ...sics.

...eigh *et al*,[29] have treated the pressure ...ations in the compression cycle by compar... the tablet with a solid (a Mohr's body), and ... cycle in Figure 6.169 is suggested. Here ... radial stress σ is plotted as a function of ... axial stress τ. The point B is interpreted ... the value where elastic recovery has its ...it, and plastic flow prevails. In a manner of ...aking this corresponds to the point P_i in Figure 6.168, but it should be stressed that the analogy is but a similarity, because the tablet mass is not nonporous.

The residual stress (AE) is the pressure exerted by the tablet on the die wall after removal of the upper punch. It follows that an equation holds

$$\tau = \mu\sigma \qquad (6.59)$$

where stresses replace forces and where μ is the frictional coefficient. One of the functions of a lubricant in a tablet is to reduce the value of μ.

The lubricant also manifests itself in how well the compression pressure is propagated through the solid mass. Tablet machines are frequently instrumented[30,31] by strain gauges or piezoelectric cells, so that the pressure exerted on the upper punch P_u and the lower punch P_1 can be monitored. The closer to unity the ratio P_1/P_u is the better the tablet is lubricated. This is obviously partly a kinetic problem, since its severity is increased with increasing speed of the tablet punches. The consolidation rate plays a part in the process, and if the time for complete consolidation does not exist, then fragmentation will take place in a structure that is not completely closely packed, and consolidation, fragmentation, and fusion will occur simultaneously.

Tablet Durability. The tablet produced must have the desired physical durability to with-

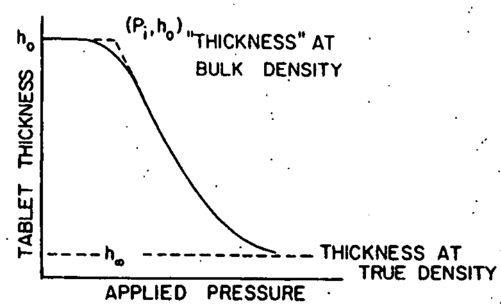


Figure 6.168. Tablet thickness h as a function of applied pressure P

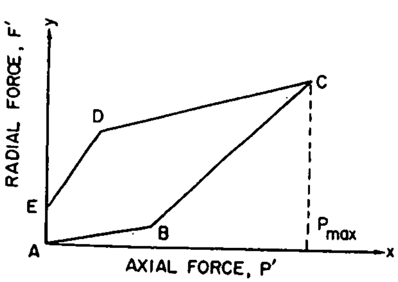


Figure 6.169. Radial force (or stress) as a function of axial force (or stress).

stand the vicissitudes of packaging, handling, and transportation. In these aspects, *hardness* is the most important quality. This is usually measured by means of a diametral hardness test: The tablet is placed (diametrally) between two anvils, and the force necessary to cause mechanical failure (breaking) is measured. This can be measured in Newton or in arbitrary units.

The systematic investigation of the diametral compression test for pharmaceutical tablets is in great part based on the studies of Newton and co-workers.[32–35] With line loading under ideal circumstances, the values of compressive, tensile, and shear stresses can be calculated by elastic theory (assuming the tablet to be a nonporous solid). The derived maximal tensile stress σ' is:[33]

$$\sigma' = \frac{2F}{\pi dh} \qquad (6.60)$$

where F is the load (Newton), d is the tablet diameter (cm), and h is the tablet thickness (cm). Some authors note that the tablet is not a nonporous solid and include a porosity term in Eq. (6.60).

The test gives different types of failure,[34] and there is a sizable scatter in results. Fell and Newton[33] have shown that the fracture strength of tablets made under identical conditions can give rise to either tensile failure (in which case the tablet splits cleanly in two parts) or to shear or compressive failure (in which case the tablet falls into many smaller parts). Newton and Stanley[34] have shown that, if limited to tensile failure, the scatter, statistically, adheres to a Weibull function.

If Eq. (6.60) were correct, then a plot of σ' versus P would be linear through the origin. The data of Fell and Newton[34] when plotted this way are quite linear, but require a small adjustment due to the nonzero intercept.

6.5.4.3 *Isostatic Pressing*[36]

General and History. Isostatic or hydrostatic pressing is a compaction of a powdered material into predetermined shapes by the application of pressure via a fluid through a flexible mold. The arrangement may be such that the flexible tool contracts or dilates by the application of the pressure. Isostatic pressing covers liquids and gases as the pressure transmitting medium, whereas hydrostatic pressing is best reserved for liquids. However, the two terms are used freely to cover both aspects. Depending on whether the flexible tool is an integral part of the press or removed from the pressure vessel after each compaction cycle one distinguishes between the "dry" and "wet bag" process.

Isostatic pressing using gases as pressure-transmitting medium is still in development and practiced by only a few. It is particularly attractive at high temperatures where compaction and sintering are combined into one operation, that is, isostatic hot pressing. The preform produced by cold isostatic pressing is in most cases further consolidated by sintering, forging, extrusion, rolling, etc. When the economics of isostatic pressing are considered, it must be in relation to the final product and not for the shaping operation alone. The advantages often lie in a better product and reduction in final machining requirement.

In 1913, Madden first described an isostatic pressing technique in a U.S. patent assigned to the Westinghouse Lamp Co; the method was developed to overcome the limitations of die-compacted billets. Madden claimed that isostatically pressed billets were uniformly compacted, devoid of strata, and possessed sufficient green strength to permit handling. Further patents were taken out on the isostatic pressing of refractory metal powders by Coolridge in 1917 (for tubes of tungsten and molybdenum), and by Pfanstiehl in 1919; Fehse described the wet bag isostatic pressing of tungsten tubes in 1928. Little further interest was shown in isostatic pressing until the 1930s and early 1940s, when a series of isostatic techniques was described by Jeffery (1932–1942) and Daubenmayer (1934) in patents assigned to the Champion Spark Plug Company.

During the same period, Fessler and Russell patented a technique for pressing spark plug insulators by direct compression isostatic pressing. These workers cited the low number of rejects, rapidity, and the need for only a limited amount of equipment as economic advantages of isostatic pressing.

By 1942, most of the advantages of isostatic pressing had been recognized, and the basic principles in common use today had been established, that is,

- The wet bag pressing of large or complex shapes in which the flexible tool is filled externally and subsequently immersed in the fluid,
- The dry bag pressing of smaller, regular shapes in which the tool forms an integral part of the pressure vessel.
- The use of rigid formers to produce accurate internal or external surfaces, and
- Pressurized by pumped systems or by direct compression with punches in a die.

Materials that had been pressed included ceramics, metals, and cermets.

In recent years, fully automatic dry bag presses for producing small ceramic components have been developed, while semiautomatric wet bag presses are used to manufacture large and sometimes complex components with reasonable dimensional accuracy and requiring only minor trimming to produce the final form. The size of pressure vessels has increased greatly. Additional materials that are now isostatically pressed include plastics (particularly PTFE), explosives, and chemicals. Isostatic pressing is also being developed for the food and pharmaceutical industries.

Hot isostatic pressing, including so-called gas pressure bonding, was developed during the last 30 years. This technique has been developed for two main research applications: the solid-state diffusion bonding of components of various metals and cermets, and the hot compaction of metal, ceramic, and cermet powders. However, hot isostatic pressing has remained confined to special applications for which the high operating costs and low rates of production are acceptable.

Isostatic Pressing Equipment. Isostatic powder compaction equipment consists of a pressure vessel, pumps to generate the necessary hydraulic pressure, and related equipment to enable effective and safe machine operation.

The time to reach the required pressure depends on a number of factors, that is, volume of the cavity, volume and compaction ratio of the powder and tool, compressibility of the fluid, and delivery rate of the pumping system. To speed up pumping, it is possible to use a number of pumps in parallel. Alternatively, a pump system using different types of pumps to reach different pressure levels may be designed.

Air-driven and hydraulically driven pumps can be built easily in a variety of modules for various demands. It is simple, therefore, to change the pumping requirements by changing the intensifier (Fig. 6.170) or increasing the number of intensifiers.

Most isostatic presses operate satisfactorily up to 400 MN/m^2 on an oil/water emulsion or hydraulic oil; for higher pressures special fluids may have to be used, but the tools used must be compatible with these liquids. Problems can arise when it is necessary to dispose of contaminated fluid after each pressing operation. Such contamination may originate

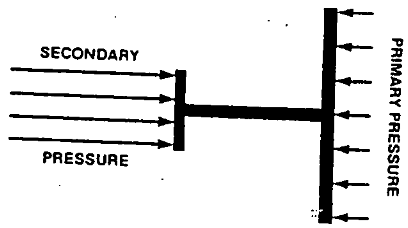


Figure 6.170. The principle of intensifiers.

from powder adhering to the external tool walls or from a tool bag failure.

To be effective, an isostatic press must be joined with equipment that fulfils some or all of the following functions: filling and consolidating the powder in the tool; loading and unloading the tool set into the vessel; handling, that is, insertion and removal of the vessel's closure; controlling the pressure in the vessel; and stripping the compact from the tool.

The difference between wet bag and dry bag pressing is illustrated in Figure 6.171. In the dry bag process, the flexible tool is fixed in the pressure vessel, and the powder can be loaded without the need to remove the tool from the vessel. The tool thus forms a membrane between fluid and powder; optionally, the tool can be placed inside a primary diaphragm so that it never comes into contact with the fluid.

Dry bag tooling is used for the production of small components at a fast rate. It is common to make provisions for loading the powder automatically into the tool by dispensing an accurately premeasured quantity. The automatic filling, the permanent location of the tool, and the smaller fluid volume result in faster operation. Dry bag tooling has also the advantage that the fluid cannot be contaminated with powder. However, because the tool has to stand up to many pressing cycles and since tool changing is time consuming, it has to be made of a very durable material.

Where mass production of simple small powder compacts (e.g., spark plug insulator blanks, grinding media, carbide tools, electrical insulators) is required, the equipment usually takes the form of a battery of small presses generally similar to and operationally having a great deal in common with a conventional hydraulic press.

It has proved relatively easy to make such a machine for automatic operation with production rates up to 90 components per minute. The compaction, ejection, and filling is demonstrated on a spark plug insulator in Figure 6.172. The production rates depend on maximum pressure, size of component, powder properties, and number of tool cavities.

The larger automatic units that have been developed include a rotating pressure chamber system where loading of powder, pressurizing, depressurizing, and unloading of the compact are carried out automatically at various work stations during the complete cycle. It has also proved possible to automatically operate certain functions on large isostatic presses. For instance, loading and unloading of the tooling, insertion and closure of the breech, pressurization/depressurization of the vessel, all have been carried out automatically by ingenious arrangements of mechanisms and controls. Figure 6.173 shows the operational sequence of a three-station rotary automatic press capable of producing parts at rates of up to 300 parts per hour.

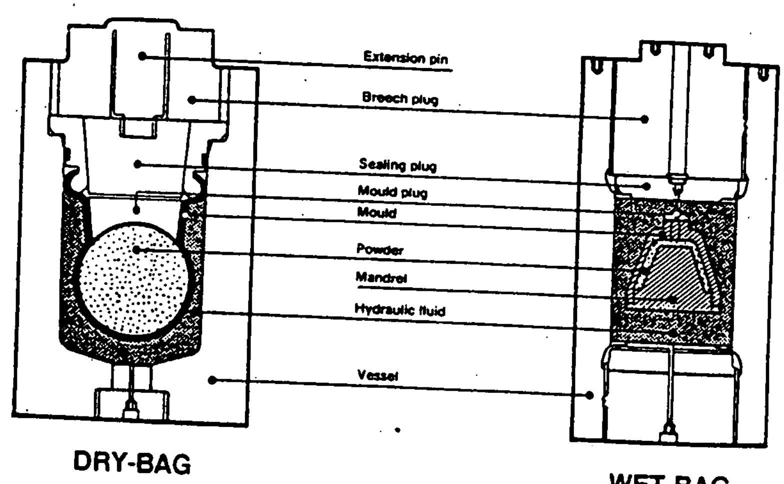


Figure 6.171. Schematic representation of the difference between dry bag and wet bag pressing.

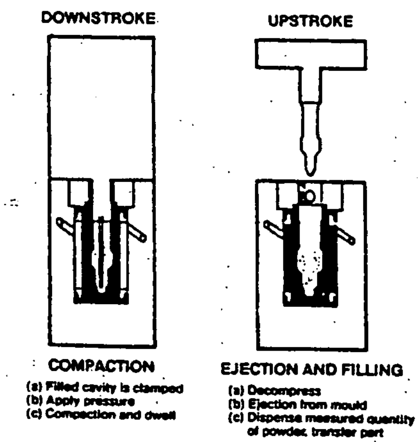


Figure 6.172. Operational sequence of a "densomatic" press (Olin Energy Systems Ltd.).

In general, the development of isostatic pressing has been comparatively slow, particularly for metal powders, and even today the technique is still regarded only as an alternative to be used when the technical limitations of conventional methods are too restrictive.

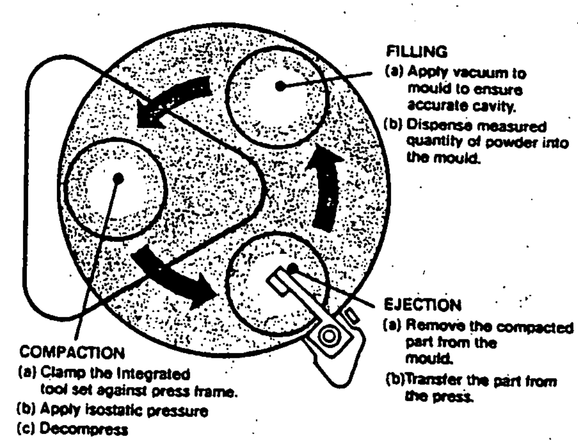


Figure 6.173. Operational sequence of an automatic, rotating isostatic press (Olin Energy Systems, Ltd).

Until recently, mostly the ceramic manufacturers have commercially exploited isostatic pressing and only to a limited extent mainly in the United States. In addition, isostatic pressing is suitable for producing high-purity ceramics and long ceramic tubes, for which there is an increasing demand. In contrast, the common metals can be readily formed by long-established methods such as casting, rolling, forging, or extrusion and only recently have metal fabricators begun to look more closely at the feasibility of isostatic pressing.

Isostatic pressing also shows great promise of becoming an established production technique for the fabrication of components from PTFE and high-molecular-weight polyethylene. PTFE, for instance, although a thermoplastic, has a very high melt viscosity, which precludes satisfactory processing by established injection moulding and extrusion techniques. This has led to the adoption of techniques used in powder metallurgy, which involve initial cold compaction and subsequent sintering, of which isostatic moulding is the latest.

6.5.4.4 Discontinuous High-Pressure Extrusion Presses

General. To illustrate discontinuous extrusion compaction of soft, formable materials with inherent or added binding characteristics, the "extrusion briquetting" process as employed by the brown coal industry shall be discussed as a typical example.

Figure 6.174 depicts the sequence of events during a briquetting cycle in a ram extrusion press.[37] The reciprocating motion is produced by, for example, an eccenter drive symbolized by the circular representation on the left. The diagram on the right indicates the progress of force exerted on the material to be briquetted. The figure is self-explanatory. Only a few important operating stages shall be pointed out.

At (3) the force exerted by the ram has reached a level that is sufficient to overcome the friction of all briquettes in the pressing channel and the backpressure caused by the column of briquettes in the cooling channel. The entire line of briquettes moves forward, with the force remaining approximately constant, and a new briquette emerges from the "mouth" of the press (4).

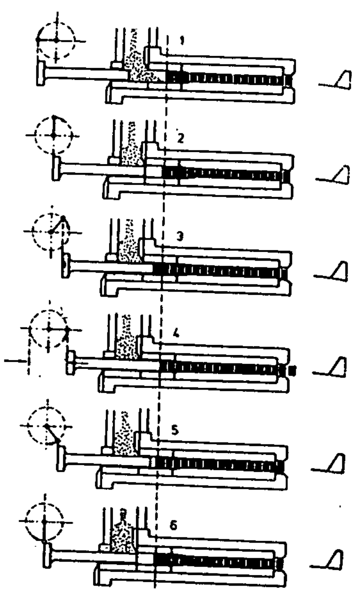


Figure 6.174. Sequence of events during a briquetting cycle in a ram extrusion press.[37]

At the beginning of the backstroke (when the eccenter drive passes position 4) at first the ram face does not separate from the briquette because of considerable elastic expansion of the briquette. It is important, however, to note that the surface produced by the ram face is so highly densified that, during the next stroke and for phases (2) and (3), it acts as the bottom of a confined volume densification chamber until friction is overcome and the product column moves forward; during the entire production sequence the surfaces of adjacent briquettes do not develop significant bonding; therefore, on discharge from the cooling channel, the product will separate into single briquettes.

Equally important is that at a typical rotational speed of the eccenter drive of 90 rpm the duration of the compression phase, during which the primary briquette is compacted, is only approx. 0.4 s.[37] Because brown coal is very elastic and the time is too short to achieve conversion of elastic into plastic volume change, the elastic recovery during the backstroke is high. Without the condition that during each compression stroke all briquettes in the pressing (extrusion) channel are again loaded and compacted, whereby more and more permanent plastic deformation is obtained, successful briquetting or organic material with high elasticity would not be possible. This is an important difference from, for example, roller presses (see also Section 6.3.3 and Fig. 6.62). That all briquettes up to the point of narrowest cross-section in the extrusion channel participate in the densification and expansion was shown by Metzner[38] and Schenke.[39]

To accomplish the above, the design of a ram extrusion press must provide a relatively long extrusion channel. However, there are physical limits to this parameter because friction and drive power as well as overall stressing of the equipment increase with channel length. Briquettes may retain a certain elastic deformation which, if suddenly released, will damage or destroy the product. Therefore, in most applications, a gradual release is provided in the channel prior to product discharge.

Figure 6.175 shows cross-sections through relatively modern ram extrusion or Exter presses. The upper channel wall is adjustable such that different release angles can be obtained. In addition, a flexible support system at this point serves as a safety device to avoid excess loading due to tramp material in the feed or overcompaction. During the backstroke the energy of the drive is stored in a fly wheel (Fig. 6.175b) and again made available during compaction.

In a closed mold, the development of a predetermined pressure presents no difficulty, but in extrusion presses the situation is complicated. The peak pressure developed at each stroke depends not only on the power exerted by the ram but also on the resistance to the forward movement of the material to be briquetted. The latter is influenced by many factors: the shape and length of the channel, die or bore, the changes in cross-section in relation to length, the smoothness of the tool walls, the nature of the material to be processed including parameters such as temperature, structure, plasticity, etc., and the type and length of the curing channel if applicable.

The rate of pressure increase is also important; it depends on stroke frequency and length and on the rather complicated relationship between movement of the ram and magnitude of the resisting frictional force between extrudate and die as well as the force caused by the column of already compressed product being pushed forward. These forces change with both state of compaction and rate of movement.

Sizing of Discontinuous Extrusion Presses. As for all pressure agglomeration methods, the most important design parameter is the compaction pressure acting upon the material to be compressed and extruded. In a machine with "parallel-wall die channel," that is, a die with constant cross-section, and without curing channel, this pressure, which is necessary to produce compacts of good quality, is determined by the static frictional resistance. It depends on the radial pressure p_r acting on the die wall, the coefficient of static friction u, and the length of the channel (Fig. 6.176).[40] Since the radial pressure and the coefficient of static friction are practically constant for a given set of conditions, channel length is the only variable for obtaining the desired compaction pressure p_K.

Recently[40] earlier theories of noncontinuous extrusion were corrected by taking into account the two distinctly different phases, that is, compression and extrusion or transport (see Fig. 6.174). As long as the compaction

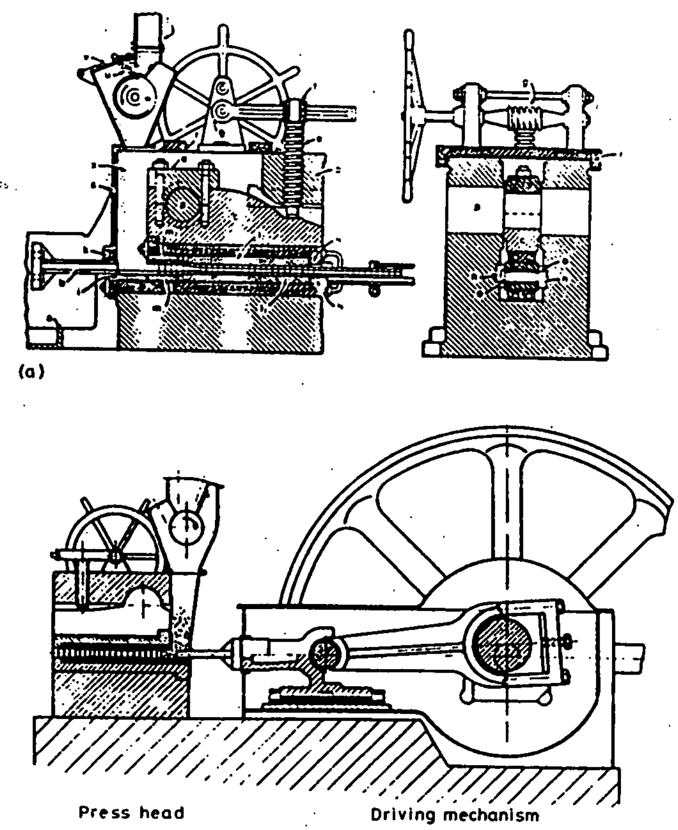


Figure 6.175. Cross-sections through relatively modern ram extrusion or "Exter" presses.[1]

pressure has not overcome the static friction of the column of already compressed compacts, the mechanism of pressure agglomeration in a ram extrusion press is the same as experienced in confined volume punch (die) presses (see Section 6.5.2). Later, during the transportation or extrusion phase, all previously densified compacts in the die are, to a certain degree, densified again while being pushed forward.

Figure 6.177 illustrates these conditions.[40] Figure 6.177a depicts the relationship between axial (compaction) pressure p_K and radial pressure p_r. $p_{r,r}$ is the residual radial pressure after separation of the ram from the elastically recovering column of compacted material in the channel during the backstroke. Figures 6.177 b.1 and b.2 show the axial and, respectively, radial pressure distributions along the length of the channel. While the axial compaction pressure drops to "zero" each time the ram retracts, a residual radial pressure always

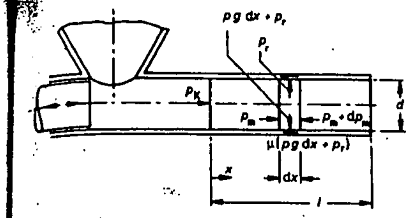


Figure 6.176. Diagram showing the development of compaction pressure in a ram extrusion press featuring a channel die with constant cross-section.[40]

remains which is primarily responsible for the back pressure p_G in the channel (Fig. 6.178) necessary to accomplish the compaction phase during the next stroke.

p_G can be calculated by:

$$p_G = p_K \cdot e^{-4\lambda\mu H/d} \qquad (6.61)$$

λ is the ratio of radial to axial pressure (Fig. 6.179):

$$\lambda = p_r/p_m = \sigma_r/\sigma_m \qquad (6.62)$$

This parameter describes the nonisotropic character of bulk particulate solids which results in the fact that pressures in the direction of loading are higher than those perpendicular to it.[41] The ratio [Eq. (6.62)] is well known from soil mechanics; is is always larger than 0 and smaller than 1. If the angle of internal friction φ and coefficient of cohesion C of the bulk material are known, λ can be calculated with:

$$\sigma_r = [\sigma_m(1 - \sin\varphi) - 2C\cos\varphi]/[1 + \sin\varphi] \qquad (6.63)$$

and the necessary length of precompressed compacts results from:

$$1 = (d/4\mu A)\ln([Ap_G/p_{r,r}] + 1) \qquad (6.64)$$

A is the slope of the de- and recompression lines in Mohr's stress diagram.[40] According to Figure 6.177b the total channel length L is $1 + H + H^* +$ densification prior to the forward movement of the column of compacts in the die, where H^* is the thickness of the new

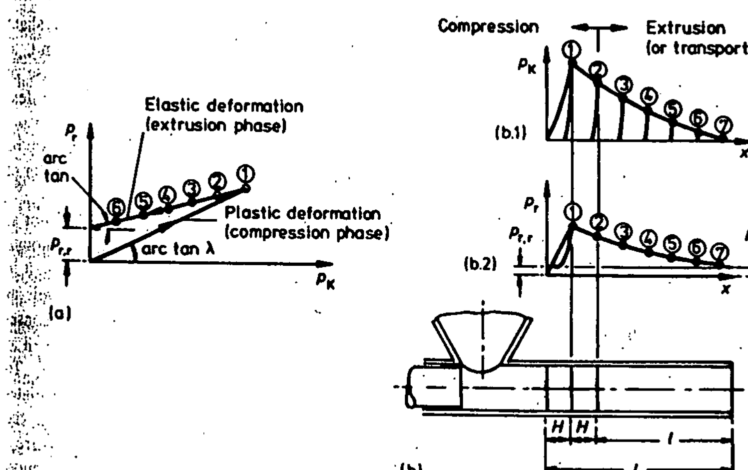


Figure 6.177. Axial and radial pressures of a compact as it moves through a channel die with parallel walls.[40]

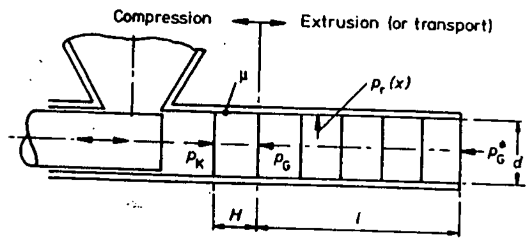


Figure 6.178. Sketch depicting the pressure acting on the particulate material in a ram extrusion press.

compact at the beginning of forward movement and H is the thickness at the dead-center turnaround point of the ram (beginning of the backstroke). Experimental investigations[40] proved that there is excellent agreement between actual data and theory.

If in additional counter pressure p_G^* acts at the press mouth onto the end of the column of compacts (Fig. 6.178), for example, because of a line of curing briquettes or a control baffle (see below) Eq. (6.64) becomes:

$$l = (d/4\mu A)\ln([p_{r,r} + Ap_G]/[p_{r,r} + Ap_G^*]) \tag{6.65}$$

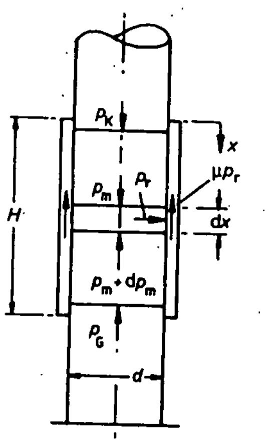


Figure 6.179. Model describing conditions in the particulate matter during the compaction phase.

From the equations a number of dimensionless parameters can be obtained that characterize the noncontinuous compression in an extrusion from open-ended dies. If these parameters are all plotted in one diagram, they can be correlated graphically which provides a method to size an extrusion press with "parallel-wall die channel."[40]

In reality, the conditions are not as simple and uniform. In most cases, the die cross-section decreases somewhat to enhance the compression phase of the method. Since this results in nonlinear differential equations, solution is not easy. Furthermore, to avoid damage of the extrudate by sudden elastic recovery when it emerges from the "press mouth" (die end), the channel walls are set at a slight taper, opening toward the discharge end, to provide for a slow and controlled release of elastic deformation. With these design features the preconditions for the above theory are no longer valid and the results can be taken to determine only approximate order of magnitude parameters.

The material characteristics are also not as constant as assumed. Relatively small inhomogeneities in the particulate solid may result in invariations in backpressure p_G as well as residual radial pressure $p_{r,r}$ and, consequently, in compaction pressure p_R as well as density or strength of the extrudate. To demonstrate the extent of variations in material characteristics, Figure 6.180 shows the compressibility presented as pressure/densification graphs of 15 lignite samples, most from the same mine and all subjected to iden-

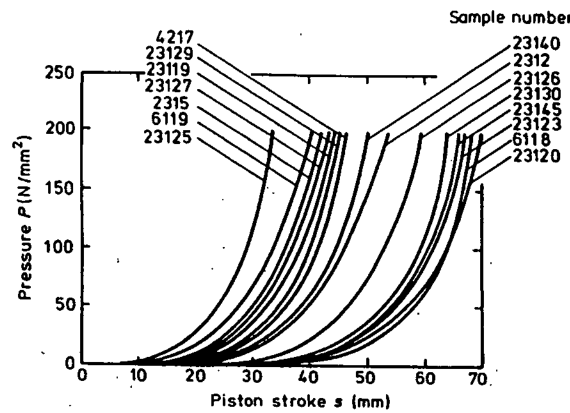


Figure 6.180. Pressure/densification graphs of 15 different lignite samples (laboratory evaluation).[42]

tical feed preparation.[42] The samples having constant weight were compacted with a maximum pressure of 200 N/mm². The large differences in compaction behavior are characterized by the piston stroke length at maximum pressure which varies from less than 35 mm to 70 mm.

There are important parameters that influence the extrusion of particulate matter. To obtain reproducible results, as many of these parameters as possible must be kept constant. The need to cool the die is rather unique for binderless briquetting of lignites in ram extrusion presses. In this application, if the die heats up, the coefficient of friction between lignite and die wall changes such that movement occurs at lower pressures, which results in less densification and inferior strength. The speed of densification, as in other high-pressure agglomeration methods, influences the amount of elastic springback. Slower speed allows conversion of a larger portion of elastic energy into plastic deformation; on the other hand, capacity is reduced by this measure.

6.6.4.5 Roll Pressing

Double Roll Presses. The most widely used roller presses are double roll presses which achieve compaction by squeezing material between two countercurrently rotating rollers (Fig. 6.181), much in the same manner as the operation of rolling mills.[43] Pockets or indentations, which have been cut into working surfaces of the rollers,[44] form briquettes or compacts.

Between smooth, fluted, corrugated, or waffled rollers, material is compacted into dense sheets. Normally, these sheets are crushed and then screened to yield a granular product.

If rows of identical pockets are machined into the working surface and the rollers are timed such that the pocket halves exactly match, so-called briquettes are formed. Roller presses do not produce compacts with the same fine detail and uniformity as those made by tabletting machines or other die presses. The flashing or web, caused by the "land areas" around each briquette pocket, which is usually found on the edges of all briquettes from roller presses cannot be removed completely and reliably and, therefore, may also be objectionable.

Because of these characteristics, roller presses find their natural field of application where relatively low investment and operating costs are more important than the absolute

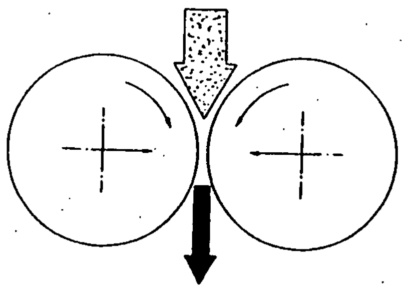


Figure 6.181. The basic principle of double roll pressing.

uniformity of the product. Double roll pressing of particulate matter is traditionally of greatest interest for all industries in which large quantities of finely divided solids, both valuable and worthless (wastes), must be handled. Originally developed as an economic method to agglomerate coal fines, today, this size enlargement technology is applied for a large number of materials in the chemical, pharmaceutical, food processing, mining, minerals, and metallurgical industries. This versatile technology lends itself to such different uses as computation and granulation of highly heat- and pressure-sensitive pharmaceutical materials, for example, pancreatin or penicillin, briquetting of extremely corrosive and poisonous materials, for example, sodium cyanide, compaction and granulation of large tonnage materials, for example, fertilizers, or briquetting of crude, hot materials, for example, metal chips and turnings, ores, or "sponge iron" at temperatures of up to 1000°C. An important, newly emerging application is the vast field of environmental control where sometimes micron or submicron sized particulate solids must be enlarged for recycling or disposal.

In the early machines and for many applications today, the particulate matter to be compacted or briquetted is fed by gravity into the nip of the rollers. Feed control is performed by adjustable tongues and distribution across the width of the roller is achieved by simple, rotating devices mounted on top of the press (Fig. 6.182).[45]

To obtain positive feed pressure and provide a more versatile means of control, screw feeders are installed for many modern applications (Fig. 6.183).[46]

The process occurring during compaction of particulate matter in roll presses is described and interpreted by different authors in a rather similar way. The feed mechanism is characterized by the pressure caused by gravity or a force fed system and the friction between material and roll surface. Compaction between two rolls may be explained by dividing the roll nip area into two zones: the feed zone and the compaction zone.

As depicted in Figure 6.184, showing a smooth roll press, the feed zone is defined by the two angles α'_E and α_E. In the feed zone, the material is drawn into the nip by friction

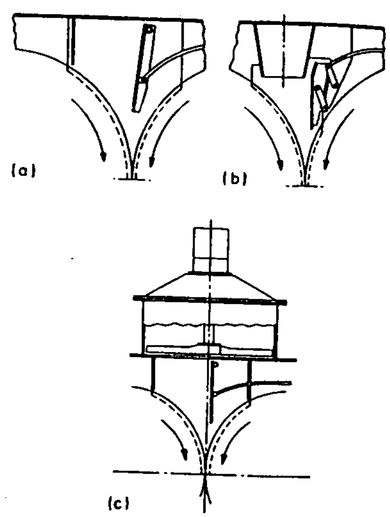


Figure 6.182. Diagrams of different gravity feed controls. (*a*) Standard tongue, (*b*) tongue with parallel movement, (*c*) mechanical distribution with standard tongue.[1]

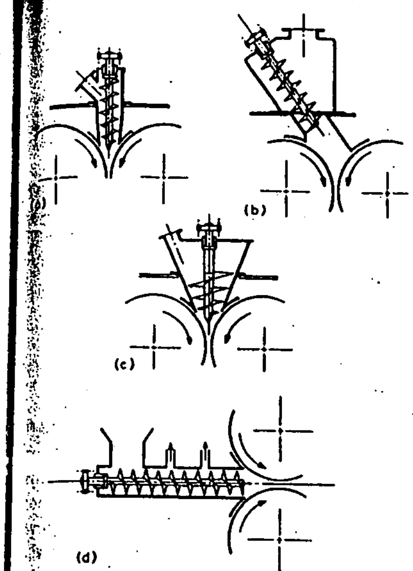


Figure 6.183. Schematic representation of some typical force (screw) feeders.[46] (*a*) Vertical straight or slightly tapered screw feeder, (*b*) inclined straight screw feeder, (*c*) vertical tapered (conical) screw feeder, (*d*) horizontal straight screw feeder.

on the roll surface. Densification is solely due to the rearrangement of particles (Fig. 6.110). The density of the feed is characterized by the bulk density ρ_0 and reaches the tap density τ_t at the point α_E. The peripheral speed w of the rolls is higher in this zone than the velocity u of the material to be compacted. α_0 is the so-called angle of delivery which is defined by the width h_0 of the feed opening above the rolls as well as the material (flowability) and feeder characteristics.

The compaction zone follows after the heavy solid line (Fig. 6.184). α_E is the angle of rolling, the gripping angle, or angle of compaction. In the compaction zone the pressing force becomes effective and the powder particles deform plastically and/or break (Fig. 6.110). α_g is the neutral angle where the sign (direction) of the friction force changes. At this point, the pressure in the material and the density have their highest values.

α_v is the angle of elastic compression of the rolls that determines the thickness h_s of the compacted sheet. α_v becomes zero and the sheet thickness h_A if the elastic deformation of the rolls can be ignored. However, in most cases the strip is even thicker than h_s owing to elastic recovery of the compacted material. The angle corresponding to this actual outlet plane is called angle of release α_R.

During compaction between essentially smooth rollers a third zone can be defined: the extrusion zone. When the direction of the friction force changes at the neutral angle α_g, the material may "accelerate" and, in respect to the roller speed, attain a higher velocity resulting in an "extrusion" through the roller gap. This phenomenon assists in the release of the compacted material from the rollers.

In the case of briquetting, the gap between the roller approaches zero and the pockets, which were cut into the roller surface and define the briquette shape, do considerably influence and change the above compaction process. Figure 6.185 depicts the mechanism of briquetting in roller presses. Of interest is only the final compaction phase. It begins when the lower axial land area passes through the line connecting the centers of the rollers. At this point, the pocket forming the briquette is practically closed at the leading (lower) edge while the trailing (upper) edge is still open and connected with the feed in the nip. Immediately following this condition the formerly closed leading edge of the pocket opens while now the upper (trailing) edge closes and compaction of the briquette is completed. Owing to "interlocking" between material in the nip and the pocketed roller surface, the previously defined feed and compaction zones are less clearly defined and determined only by interparticle friction. They no longer depend on friction between material and roller surface. However, as the leading edge of the pocket

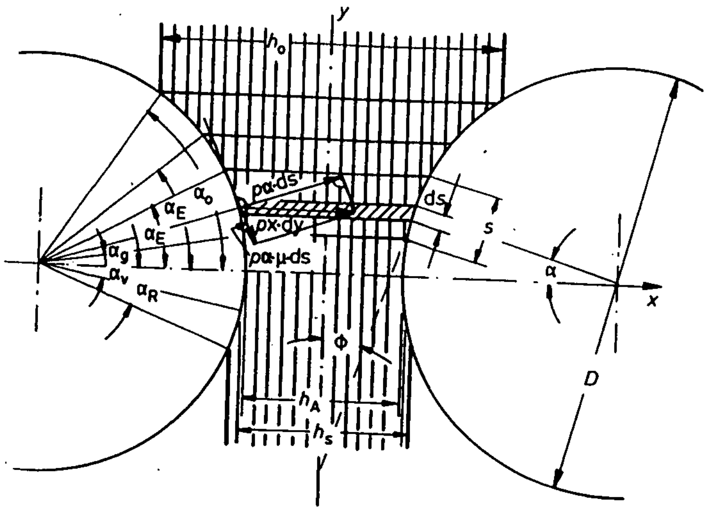


Figure 6.184. Compaction of particulate matter in the nip of a smooth roll press.

opens the force acting vertically to the line connecting the roller centers tries to "extrude" the briquette, thus assisting in the release of the briquette from the pocket, provided the shape is correctly designed.

Much of this knowledge is still phenomenological in character. A comprehensive theory of densification of particulate matter between counterrotating rollers is not yet available even though many similarities exist with the much better investigated and defined deformation of metals in rolling mills.[43]

Ring Roll Presses.[47] In the ring roll press, an alternative to the double roll press have been developed for high-pressure work. The particulate matter, normally powdered coal, is pressed between a roll and the inner surface of a ring (Fig. 6.186). Thus, a very narrow angle of entry is achieved, and with it, of course, considerable drag, which obviates forcible feeding of the powdered coal. Such a system has many advantages, but also some disadvantages that have not yet been completely overcome.

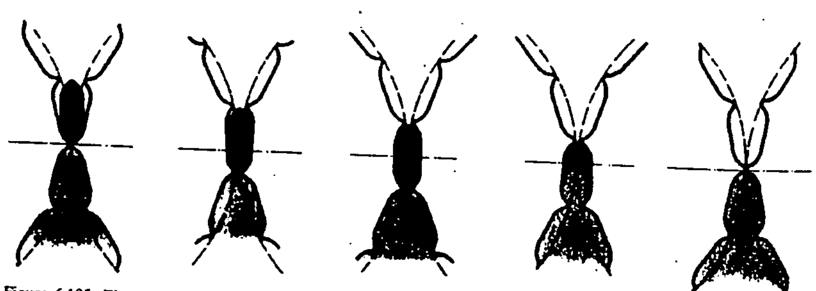


Figure 6.185. Five successive momentary conditions of briquetting between two countercurrently rotating rollers with matching pockets.[46]

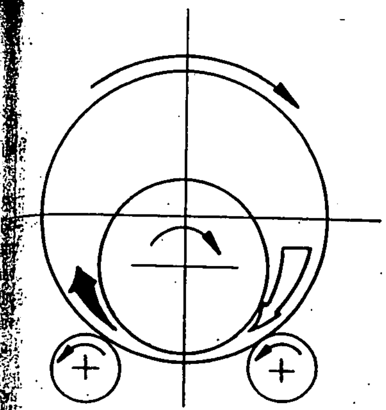


Figure 6.186. Operating principle of the ring roll press.

Sizing of Roller Presses

Theory of Rolling. The basic principle of compaction of particulate solids between two countercurrently rotating rollers (Fig. 6.187) is similar to that used in calenders for plastic foils or in rolling mills for metals. The first can be adjusted to extremely narrow gap tolerances across press rollers with face widths of up to 2 m and production speeds of approx. 100 m/min; in the latter enormous pressing forces can handle ingots of more than 35 tons weights.

While roll pressing of particulate solids is still an art rather than a science, fundamental perception and technical knowledge exist in the above mentioned fields because they were developed and investigated in modern times. Therefore, several authors concluded that it must be possible to use this knowledge and translate it into corresponding theories for roll pressing. Specifically, the basic equation obtained for rolling steel can be used to gain an understanding of roll pressing.[43,44]

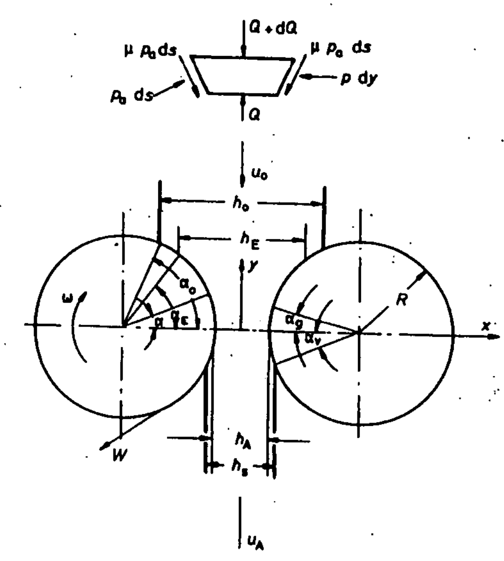


Figure 6.187. Strip model: Geometry of rolling and forces acting on a volume element.[43]

Particulate Matter. Since pressure agglomeration between countercurrently rotating rollers deals with particulate matter, results of a theory based on homogeneous and isotropic solid material is applicable only in a general way. If smooth rollers are used, a very close correlation can be obtained. Today, however, roller surfaces for agglomeration are in most cases equipped with some sort of profile to improve the "bite" on the material, which is a never ending problem because of the noncontinuity of particular matter. In case of profiled surfaces the operating zones of the "elementary theory" described by Siebel and v. Kármán[43] defining deformation and, respectively, densification cannot freely develop owing to interlocking between material and roller surface. This is most pronounced for briquetting.

The inability of the material to develop the relative speed conditions predicted by the strip model and the relatively short densification time result in considerable elastic deformation which also modifies the pressure curve as shown, for example, in Figure 6.188.[48] In the case demonstrated, a special pocket design (Koppern) is used with alternating shallow and deep cavities. Shallow pockets of one roller dip into the corresponding deep pockets in the opposite roller, much like a piston and die arrangement. The diagram at the right illustrates compaction and expansion actions and times as well as the pressure curve. The effect caused by the elastic recovery (expansion) of the briquette during release is clearly visible. This phenomenon of pressure agglomeration is very important and often determines the quality of briquetted or compacted products.

Capacity, Throughput. For the calculation of equipment capacity the macroscopic phenomenon of material passing through the nip of the roller is utilized and theoretical or actual characteristics are neglected. Therefore, the throughput C_c of a roller compactor can be determined as:

$$C_c = \pi \cdot D \cdot l \cdot h_A \cdot n \cdot 60 \cdot \gamma \qquad (6.66)$$

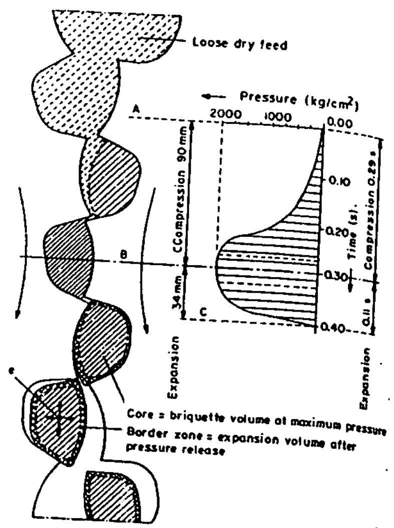


Figure 6.188. Schematic representation of the compaction process in a roll-type briquetting press[48] (1 kg/cm² = 9.81 N/m²).

where

- D = roller diameter (cm)
- l = roller length, working width (cm)
- h_A = gap width between the rollers, sheet thickness (cm)
- n = roller speed, revolutions per minute (1/min)
- γ = apparent sheet density (kg/cm³)

then: C_c = throughput of the roller compactor (kg/h).

Correspondingly, the throughput of a roll type briquetting machine C_b is:

$$C_b = z \cdot V \cdot n \cdot 60 \cdot \gamma \qquad (6.67)$$

where

- z = total number of pockets per roller, total number of briquettes per revolution

- V = volume of each briquette (cm³)
- n = roller speed, revolutions per minute (1/min)
- γ = apparent briquette density (kg/cm³)
- C_0 = throughput of the roll type briquetting machine (kg/h)

...use of leakage at the sides of the rollers ...n case of roll-type briquetting machines, ...ashings or webs around the briquettes, ...tual throughput of and the feed to roller ...es are somewhat higher (approx. 5% ...%).

...Diameter. One of the most important ...ria for the design of roller presses, which ...determines the physical size of the entire ...hine, is the roll diameter, D. It is also one ...the few parameters that is fixed in a given ...hine and cannot be adjusted to different ...ting conditions.

...eferring to Figure 6.189 it is obvious that ...sizes of the feed and compaction zones ...end on the roll diameter. Under the (al...st correct and therefore acceptable) as...ption that the gripping angle α_E changes ...ly slightly with roll diameter, the conditions of Figure 6.189 are obtained[49] for the nip ...as between two pairs of rollers with different diameters, D_1 and D_2, and identical gap, h_A. If the peripheral speed of both pairs of rollers is the same (i.e., theoretically, both machines yield identical volumetric output) compaction takes place more gradually in the case of the larger roll diameter. At the same time, a larger volume element is pulled into the nip, resulting in a higher density of the compacted product (i.e., potentially, a larger gravimetric output is obtained).

For smooth roll compactors a formula can be derived that correlates roller diameter and gap. With the definitions of Figure 6.184 and the restrictions imposed by the modified strip model [i.e., beginning at the line $h_B(\alpha_B)$ horizontal increments move with the peripheral speed of the rolls (no slip) and remain absolutely horizontal (no distortion)], the following equation for the porosity ϵ_{min} at the narrowest point ($\alpha = 0$) is obtained:[49]

$$\epsilon_{min} = 1 - \gamma_t[D(1 - \cos\alpha_E) + h_A]/\gamma h_A \qquad (6.68)$$

Since ϵ_{min} cannot become negative, it follows:

$$\gamma_t[D(1 - \cos\alpha_E) + h_A] \le \gamma h_A \qquad (6.69)$$

or:

$$h_A \ge D[1 - \cos\alpha_E]/[(\gamma/\gamma_t) - 1] \qquad (6.70)$$

In Eqs. (6.68) to (6.70) γ_t is the tap density which is assumed to be equal to the density at point α_E (see Fig. 6.184).

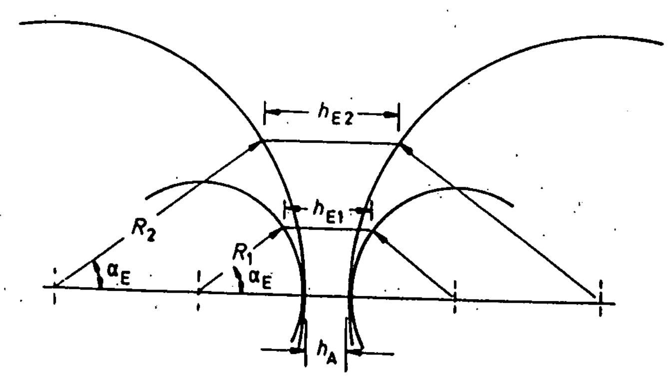


Figure 6.189. Influence of the roll di...

For materials requiring relatively little densification during compaction, the classic theory (strip model) can be used to determine the minimum roller diameter needed to form a dense sheet or briquette. Equation (6.68) can be rewritten as follows:

$$D = h_A/(1 - \cos \alpha_E)[\gamma_0/\gamma_t(1 - \epsilon) - 1] \quad (6.71)$$

where ϵ characterizes the remaining porosity at $\alpha = 0$ (disregarding elastic recovery). In the case of briquetting, an equivalent gap width h'_A must be calculated from the briquette volume and web thickness combined.

If the rollers are larger than necessary to achieve $\epsilon(\alpha = 0)$, control can be applied by restricting the flow of feed to the roll nip (see Fig. 6.182).

With increasing densification ratio the necessary roller diameter becomes larger. However, there are economic advantages in reducing the roller diameter to below the minimum diameter if materials needing high densification must be processes. Then a force feeder system must be used (see Fig. 6.183). In such a case, the above criterion can be applied to choose a diameter that is less than the dimension calculated with Eq. (6.71). The selected roller diameter should be always sufficiently smaller than the calculated minimum to allow density control by as large an adjustment of the force-feeding system as possible.

Another criterion for selection of the roller diameter, particularly of briquetting machines, is the release mechanism from the pockets (see Fig. 6.185).

Roll Gap. There is a close correlation between strip thickness h_s and theoretical roll gap h_A. Since with a given roll diameter the gap defines the compaction ratio, the strip density, γ, also depends on the gap as indicated by rewritten Eq. (6.69):

$$\gamma \sim \gamma_t[D/h_A(1 - \cos \alpha_E) + 1] \quad (6.72)$$

As a rough approximation it can be assumed that strip thickness equals roll gap. In reality, however, h_s is always greater than h_A for the following two main reasons:

1. Under load the roll gap changes because of (a) clearance in the roll shaft bearings and frame members, (b) elasticity of the machine frame, (c) deflection of rolls and shafts, and (d) elastic deformation of the roller surface.
2. After pressure release the strip expands because of (a) recovery of elastically deformed particles and (b) expansion of compressed air trapped in pores of the compact.

The extent of the roll gap change depend only on machine design and is constant for a given compaction pressure.

Expansion of the strip after pressure release is influenced by the physical characteristics of the material to be compacted (plasticity, brittleness, particle size and distribution, particle shape, etc.), the roll diameter, the speed of rotation, and the surface configuration of the rollers. With increasing roll diameter and/or decreasing speed the expansion of compacted material is reduced owing to better deaeration during densification and a more complete conversion of elastic into permanent, plastic deformation.

The smallest theoretical roll gap can be calculated using Eq. (6.70). However, because of the mechanical deformations discussed above, it is possible to roll a strip with finite thickness even if the static (= no load) gap is set at zero. This means that, in reality, the dynamic roll gap, which develops under load, must be considered. The largest acceptable roll gap results from the need to obtain a coherent compact, that is, the compaction ratio; this is influenced by the roll diameter as well as the amount and predensification of feed; the latter is characterized by the density γ_t at $h_E(\alpha_E)$. In addition, because of pressure and density gradients in the particulate mass during compaction, it is possible that the center of strip or sheet has insufficient strength if too large a thickness is desired.

For briquetting presses the correct relationship between feed rate and volume of compacts is of special importance. To avoid thick "flashings" or "webs" on the edges of briquettes, it is necessary to use strong machines, to prevent flexing, with rigid response, and a static roll gap of close to zero.

Roll Speed. For most considerations and approximations it is assumed that the peripheral speed of the rollers and the speed of the particulate matter are identical in the entire compaction zone. In reality this is not true; throughput does not increase proportionately with roll speed. The maximum speed is determined by two effects; starved conditions in the compaction zone develop if (1) too much slip occurs between rolls and material in the feed zone and/or (2) air squeezed from the particulate mass flows upward and fluidizes the feed thus reducing the supply of material to the nip.

In the first case, compaction is not high enough to form a stable compact, and intermittent operation, accompanied by sometimes severe chattering and potential equipment failure, occurs in the second case.

The minimum speed for smooth rolls is reached if the mass flow rate M_p of the free flowing powder is higher than the mass flow rate M_s of the compacted strip. Determination of minimum speed is important only if strips with tightly controlled thickness are to be made between smooth rollers, for example, in powder metallurgy. In other applications, for example, the compaction of fertilizers, the problem of minimum speed may be circumvented by selecting a narrow static gap and adjusting the hydraulic pressure such that, when feed is introduced into the nip, the clearance increases to the operating gap and, at the same time, the pressure rises to the operating level.

A completely different situation exists if the rollers are pocketed or corrugated because the flow of material is stopped when the land areas between the pockets or the ridges of the corrugations are in close proximity. For such rollers virtually no minimum speed exists. As the filling of the cups is the controlling factor and the disintegrating forces due to the release of residual elastic deformation and compressed air trapped in the pores diminish with reduced speed of compaction, briquette quality improves in most cases if the rollers are slowed down.

Roll Feeding. The simplest form of feeding roller presses is by gravity (choke feeding). A mass flow hopper with rectangular feed opening to the nip between the rollers should be used for this purpose.

The feeder dimension h_0 (see Fig. 6.184) is characterized by the angle α_0 and depends on the roller diameter, D, the gap h_A or, respectively, the surface configuration of the briquetting roll. To make use of the full transport capability of the rolls, the feed angle or angle of delivery, α_0, should be greater than the gripping angle, α_E.

In many applications the degree of compaction necessary to produce a satisfactory agglomerate is so small that the combination of commercially and conveniently sized rollers (as well as pockets, if applicable) provides too much densification if choke feeding is used. Then, the flow of material to the nip between the rollers must be deliberately restricted to avoid overcompaction (see Fig. 6.182).

In contrast, the briquetting or compacting of some other materials demands a degree of compaction that cannot be achieved by a single pass in a choke-fed roller press, irrespective of the ratio pocket size (or gap width) to roll diameter. In addition, redistribution of material (which may be extensive) from the nip against the flow of material or from the rear of cups into following cups, for example, owing to the flow of displaced air, may further reduce the efficiency of compaction. In these cases the use of force feeders (see Fig. 6.183) is required.

Roll Pressure and Torque. After determining roller diameter, width, and gap or briquette size and shape as well as roller speed using

throughput capacity and product density as input, roll force and torque as well as feed pressure must be determined. The requirements on these design parameters of a roll-type press are:

1. The press must be capable of safely supporting the roll force and sustaining the torque necessary to make a good sheet or briquette, and
2. the press with associated feed mechanism must allow development of the torque and force required to make a good product at the required throughput rate.

These parameters relate to the flow properties of the solid to be compacted.[46]

Figure 6.190 depicts schematically a typical compressibility diagram (density versus force) of a particulate solid. In a log/log plot the curve can be approximated by five straight-line segments. The first occurs at low pressures where density essentially does not change. The second range, during which density increases slowly, applies to positive force-feed systems (gravity chutes, screw feeders, etc.). The third represents the high-pressure nip region between the rollers. The compressibility factor K of the solid is characterized by the slope of the curve in this range. In the fourth segment of the curve density again remains constant; this operating condition is normally outside of the desirable working range of roller presses. In the fifth region, residual elastic deformation in the compacted solid springs back when the pressure is released.

Even though bench scale densification tests do not reliably predict the performance of a roller press, results can provide valuable information on the relative behavior of different feed materials.

The solids pressure p_{max} will be influenced by the "precompaction" pressure of the feeder, p_0. Reductions in roll force and diameter accompanying the increase in precompaction pressure lower size, weight, and cost of roller presses. In contrast, roll drive requirements remain almost unchanged[50] if the production rate is kept constant. Feed screw precompaction pressures up to and exceeding $2.8 \cdot 10^6$ Nm^{-2} have been reported. In normal operation the pressures are probably in the range of 10^4 to 10^6 Nm^{-2}.

Feed screws are axial flow type compressors whose power requirements increase with the compression ratio and also with larger frictional forces between material and screw occurring at the higher pressures provided, however, that the permeability of the densifying bulk mass remains high enough to allow unrestricted flow of the gas that is expelled during compaction. The total power requirement of the roller press with screw feeder is the sum of both drive energies. Figure 6.191 illustrates schematically the correlation between total drive energy and precompaction pressure for an always highly permeable particulate solid and for a material with decreasing permeability during compaction. Normally, the share of feed screw power in relation to total drive power is the range of 1% to 20%.

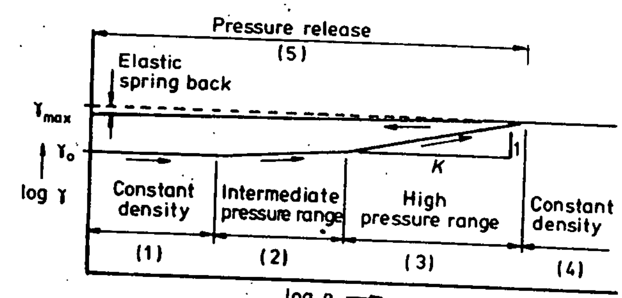


Figure 6.190. Typical compressibility diagram (density versus force) of a particulate solid.[46]

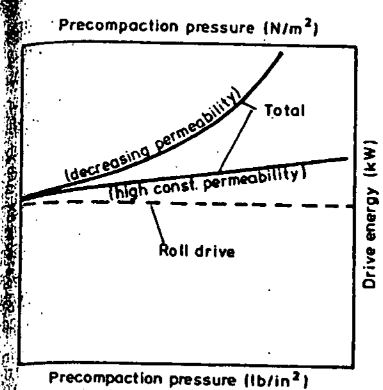


Figure 6.191. Drive energy of roller presses with screw feeder as a function of precompaction pressure and influence of permeability during compaction.

Optimum precompaction pressures, p_0, and corresponding feed screw designs vary widely with physical and chemical properties of the material and also with the desired quality and shape of the material. Because of the many different applications and the numerous variables, optimum precompaction pressure and feed screw design are determined during tests with a sample of the actual feed material whereby roller presses with large roll diameters are often used to avoid scale-up problems and alternative feeder designs are applied. Actual plant conditions are simulated by adding the proper amount of "recycle" to the feed.

Scale-Up Considerations. In addition to the above considerations, there are some simple relationships between roll diameter, D, force or pressure, p, and gap width, h_A, which can be applied for scaling-up or -down. An equivalent gap width may be calculated for briquetting machines and used as approximation.

These relationships are:

$$\sqrt{D_2/D_1} = p_2/p_1 \qquad (6.73)$$

$$\sqrt{h_{A2}/h_{A1}} = p_2/p_1 \qquad (6.74)$$

Although it follows from Eq. (6.73) = Eq. (6.74) that the sheet thickness (i.e., gap width h_A) can be larger with increasing roller diameter, experience teaches that the prediction $h_{A2} = (D_2/D_1) \cdot h_{A1}$ can normally not be achieved. Depending on the characteristics of the material to be compacted the minimum sheet thickness may be estimated during scale-up by:

$$h_{A1}(D_2/D_1) > h_{A2} > \sqrt{D_2/D_1}\, h_{A1} \qquad (6.75)$$

Special Characteristics of Roller Presses

Phenomenology of Roll Compaction. Controlled and complete removal of gas (normally air) compaction that is expelled during densification is an important consideration for all pressure agglomeration methods. Correct and sufficient venting becomes most critical for roller presses handling large bulk volumes. For example, during roll compaction, densification ratios are typically 2:1. In the case of potash compaction, a common high-capacity application of roller presses, the bulk density of the feed is approx. 1 t/m^3; it increases to an apparent density of the compacted sheet of nearly 2 t/m^3; therefore, approx. 0.5 m^3 of air per ton of salt must be removed during compaction. Since modern, large-scale equipment is capable of handling approx. 80 to 100 t/h, 40 to 50 m^3/h of air is to be vented without disrupting uniform operation of the press.

In many applications the simple smooth, cylindrical roll surface design is used. Particularly with smaller roller diameters the gripping angle of compaction, α_B, becomes very small, resulting in reduced compaction ratio, and, therefore, a lower throughput. Especially if fine powders are to be compacted, a force feeder is necessary to overcome these shortcomings. A rough surface will increase the gripping angle and improve the situation; however, because of inevitable wear, which will

eventually polish the surface, this measure is only of limited value.

Other surface "irregularities" include different types of corrugations or shallow pockets that produce "waffled" sheets.[51] The latter seem to improve deaeration.[52]

While profiled rollers are acceptable for some small-capacity applications and for materials with low abrasivity, large, high-capacity machines processing abrasive solids require the inherent advantages of smooth rollers, which are: rugged design, easy manufacturing, the possibility of refacing work rolls, and lower price. To improve the angle of grip, weld beads may be applied to the surface; these welds can be replaced from time to time as required.

A relatively homogeneous sheet is most easily formed across almost the entire width of the rollers with small roller diameter or narrow roll gap (sheet thickness) and low circumferential speed (both resulting in small capacity) as well as small compaction ratio. Then, only a relatively small amount of air is expelled which can escape partly to the top and partly to the sides of the rollers. Production of a homogeneous sheet will be further facilitated by coarser feed and correspondingly larger pore diameters (permeability).

Figure 6.192 illustrates the forces at work in a roll press when powders are compacted.[53] The rotating rollers magnify the small contact pressure between the solid particles from a value correspondingly to p_0 at the feed point of the press to the maximum pressure, characterized by p_{max} near the narrowest point of the nip (part A of Fig. 6.192). The increase may be calculated as a function of roll diameter, sheet thickness, and roll friction coefficient as well as the material's compactibility, interparticle friction, and permeability. On discharge, the pressure is released and initial strength is caused by binding mechanisms that have been activated during compaction. Residual elastic deformation at the point of discharge is relieved by expansion of the sheet which may result in a weakening of the binding forces.

Part B of Figure 6.192 shows the corresponding increase in apparent density which is typically approx. two times higher after compaction but may reach values of up to three times the feed density depending on material as well as roll press size and design. As can be seen, densification occurs very rapidly (in 1 s or less) in the narrow part of the nip. Ideally the compact density remains constant after discharge but may become somewhat lower due to elastic expansion (dotted curve).

Part C of Figure 6.192 shows that, depending on permeability of the particulate solids being processed and its change during compaction, respectively, air pressure in the material may increase to different levels. If residual porosity (permeability) remains high enough during densification and in the compacted sheet, this air pressure equalizes by gas flow

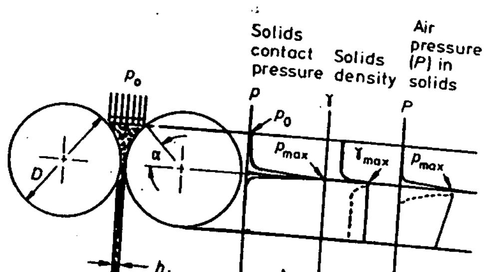


Figure 6.192. Representation of forces at work in a roll press when powders are being compacted.[53]

venting both during and after compaction. ermeability is or becomes low, air pressure ases to very high levels and the sheet nds on release similarly to the effect of tic recovery mentioned above. However, use it is not a relaxation of the material f but an "explosive relief" of compressed located in the interstices (pores) between solids, expansion of the sheet by this hanism always results in some, often serious destruction (bursting) of the compact. sting is frequently associated with a pop noise. Sheets may break into slivers or gularly shaped pieces and sometimes disinrate to powder.

he damage that is done by the expanding depends on the strength of the compacted erial (Fig. 6.193) and its remaining open rosity but there is always some reduction of ality associated with the process. Therefore, it is most desirable to remove the air while it is pelled from the densifying material. If initially relatively narrow rollers, arranged side by side, are considered, three types of problems created by the removal of entrained air can be identified.

First, gas flowing countercurrently to the feed material in the nip causes particulate solids to alternatively fluidize and flow. These process conditions repeat in a cyclic manner. This operating condition is not acceptable because not only does the yield of good product drop considerably, thus reducing process economics, but equally important is that large fluctuations in pressure and torque are experienced that may result in serious damage to rolls, bearings, gear reduces, and drives (chattering).

Second, the gas flowing into higher levels of a gravity feeder hinders the free flow into the nip and reduces the roller press capacity. This problem can be overcome by installation of a feeder (e.g., screw feeder) which forces material into the nip.

Third, the conditions described above are most pronounced for fine feed materials featuring low permeability. It is possible that the problems caused by entrained air cannot be solved by simple and economical (i.e., sufficient roller speed and, thus, capacity) means without changing the feed characteristics by coarsening the particle size and, therefore, increasing permeability. One rather simple method to achieve this is to recirculate a certain amount of crushed compacted material with a particle size distribution that must be determined by experimentation.

Figure 6.194 shows the effects of roller speed and permeability on air pressure in the compacted sheet.[53] To the original graph a line has been added that characterizes the theoretical strength of the compacted sheet (i.e., prior to decompression); it decreases with increasing roll speed because of the shorter time available for the development of binding forces. Comparison of this line with the curves for air pressure shows that air entrainment does not limit roller speed for coarse granular

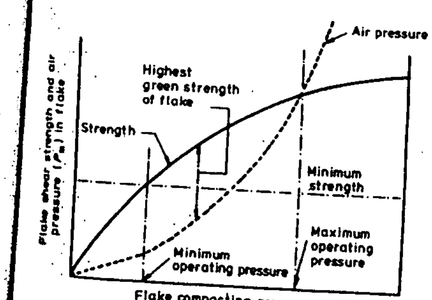


Figure 6.193. Effect of entrained air on compacted sheet strength.[53]

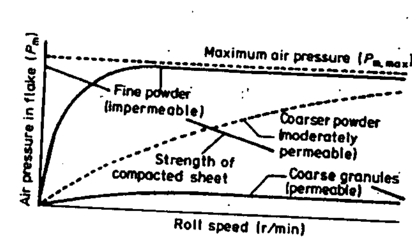


Figure 6.194. Eff

material, and only insignificantly influences the choice of roller speeds for "moderately permeable" coarser powder, but leaves only a small range of very low speeds for "impermeable" fine powders.

In most cases, the feed of roller presses does not consist of the coarse granular material with no limitation to roller speed. Consequently, if equipment with large capacity is required, roller width must be increased. Figure 6.195 reiterates[52] that air can escape from the nip countercurrently to the flow of material into the feeder arrangement, over the top of the rollers, and sideways between the cheek plates sealing the roller nip against excessive leakage of solids. The first portion, which causes limitations of free flow of feed to the rollers, grows with increasing roller width. While wide rollers (with working widths in excess of 1000 mm) operate without problems in high-capacity applications if materials with "high permeability" are handled,[52] decreasing feed permeabilities will reduce acceptibility of wide rollers, even if force feeders are applied.

Generally, the same phenomenon as discussed above occur during briquetting with roller presses. Differences are, that it is more difficult to vent the gas that is being squeezed out from a pocketed roll, particularly during the last stages of compaction when the pockets close (see Fig. 6.185) and essentially seal remaining air within the briquette. Since this compression of residual air cannot be completely avoided even at low roller speed and high permeability of the particulate solids and, on the other hand, during briquetting a final product is to be made, the disruptive effects of entrained air are even more critical during and after release from the rollers than in the case of compaction.

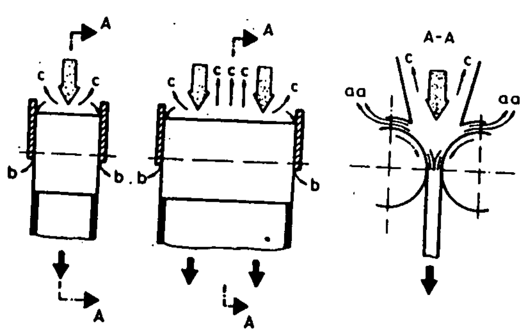


Figure 6.195. Schematic representation of deaeration in a roller press.[52]

Phenomenology of Roll Briquetting. During roll briquetting individual pieces with defined shape are generated but are not compacted simultaneously all over; rather, pressing takes place at varying rates and reaches different maxima at different times in separated points within the briquette. Only in the relatively rare case of materials with a very high intrinsic bond strength caused by compaction and requiring a low degree of densification can the product of roll briquetting be described as fault-free. Even in these cases the compact is not a perfect match to the pockets. The generative process of rolling always produces a compact that is longer than the circumferential length of the cup. This process, together with expansion due to elastic recovery and/or compressed air make the briquettes larger than the combined pocket volumes. If other materials are briquetted, especially those requiring high densification, imperfections and faults do arise that may not occur in every compact and, often, very similar problems can arise for entirely different reasons. Moreover, the precise causes of some of these faults are still unknown.

One of the most easily recognized and probably the best understood of the various faults is a narrow, broken band of material around the plane dividing the two briquette halves. This is commonly known as "flash" or "web" and results from the fact that the rollers are not in contact during operation. The web can become excessively thick owing to either stretch of the press frame or misalignment of the rollers during the setting-up procedure; in that case, briquettes are joined together and, particularly in the case of multirow presses, may have the appearance of a chocolate bar. In addition to distracting from the appearance of the product, special equipment is necessary to separate the briquettes which may also cause damage to the structure.

Another fault, equally as common as that of thick flash but probably less understood, is that in which the compacts open up along the plane of pocket contact. In the vast majority of cases, this opening is at the trailing (last compacted) edge of the briquettes but, occasionally, opening at the leading (first compacted) edge has been described. These faults are known as "clam-shelling," "oyster-mouthing," or "duck-billing."

The most common explanation of the above, especially with low-plasticity materials, is that, in attempting to achieve adequate compaction at the leading edge, the trailing edge is subjected to excessive pressure and also contains most of the compressed air; therefore, it splits as a result of elastic recovery and expansion of air when the briquette is released. However, as the phenomenon has also been observed for very plastic materials in which even forward extrusion has occurred, it is likely that other mechanisms participate in producing this fault. Breaking away the flash may be a source of cracks which could lead to splitting along the central plane. This would also provide a satisfactory explanation for clam-shelling, at the

Because the trailing edge of briquettes does not receive its final pressing until the front ends of the pockets have separated (see Fig. 6.185), compacts are not homogeneous in density and, in general, using a symmetrical cup shape, the trailing end is distinctly denser than the leading end. This may suggest that the rear end undergoes higher rolling load than does the front; this, however, is not always the case. The difference in density is least when the material is plastic because it will flow, both in part and in whole, and may even extrude forward when the cups open at the leading edge. Such flow can also result in a highly polished surface of some finished briquettes.

A near uniform state of stress and strain within a briquette is more difficult to achieve with a roll press than with uniaxial compaction presses (either closed mold or extrusion) because of the more complicated geometry of the "pressing chamber" (nip plus briquette pockets). Homogeneity (but not necessarily isotropy) could be attained if either:

1. A cup could be designed that would apply equal strain increments to all elements of the material without gross movement of the materials within the cups, or
2. the material is deliberately made sufficiently plastic (either by previous processing or the addition of a plasticizing constituent) to allow equalization of strain throughout the material during compaction.

Neither of these extreme situations is feasible. For case (1) no practically conceivable cup shape can produce equal strain increments; and in case (2) a material with the necessary degree of plasticity will normally be incompatible with a potential need to develop adequate pressing load because the material could be extruded from between the pockets at relatively low pressure. Alternatively, the product specification may exclude modification of the material or it is impossible to remove the plasticizing constituents after briquetting if they are inacceptable in the product. However,

material featuring maximum plasticity commensurate with required pressing load and product specification is likely to give optimum briquette equality.

One factor that may contribute to the nonuniformity of strain is an increase in roller speed. During the main stage of the compaction process, the strain rate in volume elements varies from point to point in a cup and with cup position. In the simplest geometrical estimation of these strains, their rates of change will be directly proportional to roller speed. Therefore, it is likely that operating a briquetting roller press at the slowest possible speed consistent with economic throughput would be advantageous in reducing stress differences during compaction. Moreover a slower roller speed will allow more time for any time-dependent recovery to attain equilibrium and plastic flow to reduce high stress concentrations.

Extraction Considerations in Optimizing Pocket Design. Equally as important as designing a pocket shape to achieve stress-free compaction is the requirement to obtain stress-free extraction. Even if the briquette experiences a fairly uniform stress distribution at the point of minimum volume (owing to a combination of optimum pocket shape and good material characteristics) and is, at this point, relatively fault-free, it can be damaged during its release. Although the release portion of the cycle is geometrically the same as the compaction portion, the material has changed from a deformable particulate solid to a coherent mass that is often under considerable elastic deformation. Consequently, the principal release problems are associated with changing stress distribution within the compact.

Because the trailing edge of the briquette must ultimately attain a near closed shape, with the lands at the rear of the pockets almost touching, the rolls will continue to apply pressure until the land between successive cups passes the plane of roll axes. During this phase, the forward cup space is already increasing in volume and the constraints to the leading part of the briquette are released while the back is still being compacted (Fig. 6.185).

The effect of this mechanism will be between two extremes: one for a highly elastic low modulus material and the other for a completely inelastic (or very high modulus) material.

Briquettes made from elastic materials can always expand sufficiently at their leading end to support the rear stress during the critical period and, except in the unlikely case that the new stress distribution is so distorted that briquette strength is exceeded at some point, the compact will remain undamaged. In contrast, inelastic briquettes cannot follow the receding pocket surfaces by expansion; therefore, it moves forward until the front edge protrudes beyond the plane containing the receding edges of the cup and very high stresses can be generated at the line or point contacts with the compact. Some damage to the briquette is almost inevitable. Furthermore, the trailing edge of the compact may remain comparatively weak because not enough material is contained to fill the now larger briquette volume. If the material can deform plastically extrusion of a "tongue" through the opening gap into the rear of the preceding compact may occur.

Secondary release problems arise from various adhesive forces between briquette and cup. Obviously, pockets cannot contain any reentrant surface because, as the pockets part, the briquette would get caught and tend to split in half. Similar forces can be caused by friction between briquette and cup and on surfaces nearly parallel to the roller radius (Fig. 6.196, left).

Generally, three factors must be considered in optimizing the pocket shape for easy release of briquettes:

1. The overall release geometry. This is governed mainly by the ratio "roll diameter/pocket length." If this ratio is large enough, the trailing edges of the cups will close before the leading edges have separated sufficiently to cause damage or extrusion.

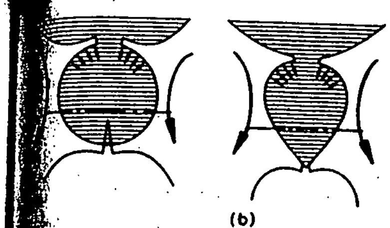


Fig. 6.196. Schematic representation of two extreme pocket shapes. (*a*) Half circle, no briquette production possible owing to release difficulties; (*b*) "rationally designed" pocket (tear drop).

2. The detailed release geometry. This is governed by the pocket shape. A "pillow shape" with the axis of its partial cylinder across the rollers and conical sides of wide angle will probably be the best. It is suggested[46] that at no point on the cup surface the normal to the surface should differ in the direction from the roll radius by more than 65°.

3. The properties of the material as briquetted. Pocket design will be more critical for inelastic than for low-modulus compacted material. However, the design will be less critical if the compacted material features high shear strength. If the front part of the briquette can survive the high stress resulting from the rear load because of its inherent strength, then any sophisticated cup shape compensation is unnecessary.

In many actual cases, compact shape must conform to commercial requirements that are unrelated to the production process (e.g., a distinctive shape may be desired for a proprietary fuel or special identifying marks may have to be applied). Consequently, the cup shape used may not necessarily be the optimum design for the material to the compacted.

The Difference in Behavior Between Single with Multirow Briquetting Presses. The different behavior of wide roll compactors as compared to narrow rollers has been discussed.

Roll type briquetting presses feature even more pronounced differences if single row designs are compared with multirow applications (i.e., two or more pockets across the face of rollers).

For most materials, the throughput of a single row press can be increased by placing two or more rows of pockets side by side on the (correspondingly wider) rolls and enclosing the space with a single pair of cheek plates. Theoretically, the limitations of this method of increasing throughput are only in the need to provide adequately sized bearings to support the increasing roll load and in distributing feed uniformly between all rows.

Although briquettes made in a single row pilot plant may be of excellent quality, for some materials performance of a commercial multirow press may be unsatisfactory. Such problems are normally encountered with materials demanding high compaction ratios.

Three main reasons may explain the operating difference between single and multirow presses:

1. It has been noted that proportionately more work is done in the precompaction stage of a single row press. This extra work may result in a general degradation of feed material, change of the position at which compaction begins, increase of the bulk density at the start of compaction, even a difference in the adhesive properties of material's surface.
2. In the case of multirow presses it may be impossible to achieve an adequately uniform distribution of the feed on the rollers. Part of the maldistribution may be due to uneven gas backflow, particularly in the center of wide rolls. The influence of uneven distribution becomes more critical as the briquette volume decreases. With very small pockets it becomes almost impossible to produce briquettes of uniform quality in multirow presses.
3. In single row presses the cheek plates may

bution of material within the cups. The distribution within the pockets is more critical in systems requiring high compaction ratios. The effect of cheek plates is less pronounced or absent in multirow presses.

Entrainment of Material by Roller Presses. The mechanisms that control the entrainment and subsequent movement during densification are not yet fully understood. However, a number of theoretical approaches have been successful in predicting the behavior of roller systems, particularly if small changes in density are involved.

Originally, most workers considered a horizontal volume element of material in a roll press with rollers arranged side by side and assumed that it remains horizontal and retains constant thickness as it moves through the nip between the rolls. This is a gross oversimplification and leads to the prediction of excessively large changes in density for a given roll system if the material is "entrained" at the angle of friction.

Therefore, later research concluded[54–58] that material is entrained at some other angle —the "true angle of nip"—which is smaller than the angle of friction and must be determined experimentally. The use of an empirical "angle of entrainment" makes allowances for the "upward movement" of material avoiding the squeeze after compaction has commenced.

For additional information on roller presses, particularly special design features, instrumentation, and control, as well as peripheral equipment for systems with roller presses, the available literature should be consulted.[1,43,46,59]

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6.6 OTHER AGGLOMERATION METHODS

6.6.1 General

[redacted] Different techniques use different binding mechanisms and the equipment applied to accomplish agglomeration is characterized by suitable handling and treatment of particulate matter to bring about the desired effect. For example, in tumble agglomeration, the particulate solids are subjected to movement that is irregular, often turbulent, and controllable, resulting in collisions between particles, development of bonds, and growth of agglomerates. In pressure agglomeration a more or less stationary bed of particles is consolidated by pressure bringing about various binding mechanisms.

[redacted] Consequently, "other" agglomeration methods still employ similar effects and mechanisms as mentioned before in the two main groups: tumble (Section 6.4) and pressure (Section 6.5) agglomeration. Most of the examples that will be discussed in the following are intended to show that for special applications and tasks knowledge of the binding mechanisms as well as creativity in regard to techniques to be used may result in special new methods for solving a particular problem more economically or conveniently than currently available through existing technologies.

6.6.2 Agglomeration Heat

Agglomeration by heat uses primarily the binding mechanisms sinter (or mineral) bridges and partial melting (Fig. 6.2). It is frequently called "sintering."

In the first edition of this book Limons[1] covered the sintering of iron ores in much detail; this treatice is recommended as a ready reference.

Often, agglomeration by heat is a second step (curing) in an agglomeration process, whereby in the first stage size enlargement to discrete agglomerates occurs by means of tumble or pressure agglomeration methods with or without binders and, in the second stage, hardening and development of permanent bonds is achieved by heat.

The largest application of such two-stage agglomeration procedures is the pelletization of iron ores.[2-8] Figure 6.197 shows schematically the three main induration methods used in this industry.[2] They are the vertical shaft furnace (a), the straight or sometimes circular (traveling) grate or strand machine (b), and (c) the combination of straight grate and rotary kiln ("grate-kiln"). In a complete pelletizing system these induration methods are combined with tumble agglomeration in drums or discs.

The final, often very high strength of agglomerates is obtained by development of solid bridges between the ore particles at elevated, so-called "sintering" temperatures. In the first, the tumble agglomeration stage, nearly spherical pellets are produced. These "green" agglomerates are held together by surface tension and capillary forces. During induration the pellets must be first dried and preheated before, at approx. two thirds of the melting temperature, migration of atoms and molecules sets in at solid/solid interfaces and solid bridges are formed. The problem with this and many similar processes is that, after drying, the original binding mechanism of the green agglomerates (capillary forces and surface tension) has disappeared but sintering has not yet begun. Therefore, there is a time during the process at which the agglomerates exhibit almost no strength. Theoretically, only the traveling grate may introduce low enough stresses into the

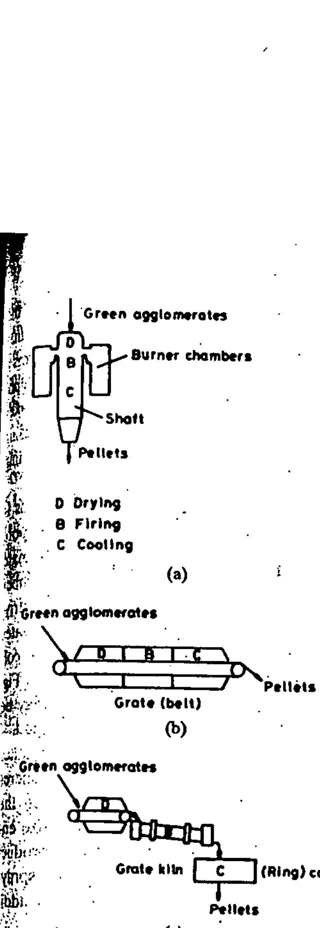

Figure 6.197. Schematic representation of the three major induration methods used in iron ore pelletization. (*a*) Shaft furnace, (*b*) grate, (*c*) "grate-kiln."

pellets that the latter can survive this phase. In reality, even these machines, because of their relatively crude design, have vibrations and other dynamic forces that endanger survival of the weak agglomerates. To overcome this problem, additives are used during tumble agglomeration that retain some bonding characteristics in the dry state and improve the change of survival until sintering begins. In iron ore pelletizing, this additive is traditionally bentonite, a natural montmorillonite clay.[9] In the wet stage this material imparts plasticity and in the dry stage some, but sufficient strength. Unfortunately, the addition of bentonite not only increases the cost of pelletizing but also introduces impurities (slag components). Therefore, recent efforts to optimize the process have come up with organic additives[10,11] that do retain strength in the dry stage but burn out during sintering, thus preventing unwanted contamination.

The principle of first forming and then indurating agglomerates is also applied for other materials, particularly nonferrous ores and metal bearing recycled or reclaimed wastes.[6]

Sintering as a process of solidifying and densifying powders is very often used in modern powder metallurgy and for manufacturing of high-quality technical ceramics as well as composite materials, for example, cermets. Because of the need for good control of the process and extreme final quality of the products, a theory of sintering has been developed for these applications and extensive research has been carried out.[12-16] During sintering, shrinkage takes place that is correlated to the density of the "green" (preagglomerated) part. Since most "preforms" are produced by pressure agglomeration, density gradients (see Fig. 6.111) can cause distortion during sintering. It is therefore most important to select the correct tooling (see Section 6.5.4.1). To obtain small density variations and little distortion, isostatic pressing may be used for the production of agglomerated preforms (see Section 6.5.4.3).

6.6.3 Spray Solidification

Several methods are known that convert droplets, formed from a melt, into solid granular products by cooling. These processes are called prilling, spray cooling, spray solidification, spray congealing, as well as shoe or pastille forming.[17] Although these methods are often mentioned in connection with agglomeration, this technology is not part of the unit operation "Size Enlargement by Agglomeration."

Spray drying (Fig. 6.198) on the other hand, is a true agglomeration process. Feed material is either a solution, an emulsion, a suspension, or a slurry. While in the first case particles